IVG INSTITUT FÜR VERBRENNUNG UND GASDYNAMIK

UNIVERSITÄT DUISBURG ESSEN

International Bunsen Discussion Meeting and Workshop on

Laser-induced incandescence: Quantitative interpretation, modelling, application

September 25-28, 2005

Universität Duisburg-Essen, Germany

Organizers:

Christof Schulz
IVG, Universität Duisburg-Essen
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Co-organized with:

DBG SFB445

International Bunsen Discussion Meeting and Workshop

Laser-induced incandescence: Quantitative interpretation, modelling, application

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UNIVERSITÄT DUISBURG ESSEN

Laser-induced incandescence (LII) has proven to be a powerful tool for particle concentration and particle size measurements in combustion, particle synthesis, as well as in environmental applications. Different experimental approaches and data evaluation techniques exist. However, the links between the various groups that are currently working on making LII truly quantitative seem to be quite weak. In order to strengthen the community, the status of the current understanding regarding LII diagnostics will be discussed at this joint meeting. The opportunity exists to gather the leading researchers in the field for a forum which will identify weaknesses and knowledge gaps and will provide perspective to focus future LII research. The outcome of this workshop will be of great assistance in determining the possibilities and limits of LII, and will provide the initiative for further research on enhancing the fundamental understanding of LII.
The meeting combines conference-type presentations on the first day with a workshop on the second and third day. Attendees of the workshop have been encouraged to apply their experimental techniques to pre-defined measurement situations before the meeting and to apply their data evaluation techniques (model, calculation) to sample data that will be made available well in advance of the meeting. Results will be compared and discussed during the workshop. The two parts will be structured in the following way:

Conference (Monday, Sept. 26)

The conference presents a wide variety of new aspects of LII. It does *not* cover *applications* of standard LII techniques. Topics are: Combinative measurements, non-carbonaceous materials, measurements in extreme situations, determination of material constants, and other novel developments related to LII. Full papers can be submitted by Dec. 31, 2005 to appear in a special feature in Applied Physics B.

Workshop (Tuesday and Wednesday, Sept. 27-28)

Key areas of experimental techniques, modelling approaches, material properties assumptions as well as the outcome of the joint effort on pre-defined measurement conditions and sample data evaluation will be discussed. The different topics have been assigned to topic leaders. Workshop attendees have been encouraged to send material to the topic leaders. The topic leaders will compile the different views and present them to the general audience with time for discussion. The presentation and discussion time, however, will be limited in order to be able to cover the complete field without parallel sessions. Topics that show further need for discussion will be continued on Wednesday morning in split sessions. The results of the group work will finally be presented to the general audience in the afternoon sessions. If possible, agreement on standard models and standard material properties should be sought. The results will be made available after the conference on a website devoted to LII research. The topic leaders will put together the material and discussion points in respective chapters of a conference report that also should be published as a journal article.

Website

www.uni-duisburg.de/ivg/vg/lii-workshop

Contact

e-mail: office@ivg.uni-duisburg.de fax: +49 (0)203 379 3087

Organizers

Christof Schulz (IVG, Universität Duisburg-Essen)
Greg Smallwood (NRC Canada, Ottawa)
Bas Bougie (Radboud University Nijmegen)
Coralie Schoemaecker (Université des Sciences et Technologies de Lille)
The meeting will be co-organized by the Bunsengesellschaft für Physikalische Chemie and by Sonderforschungsbereich 445 “Nanopartikel aus der Gasphase”, (Universität Duisburg-Essen)

Location

Haus der Unternehmer, Duisburg
Düsseldorfer Landstraße 7
47249 Duisburg
Telephone +49 (0)203 60820
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Registration

Registration is possible during the get together on Sunday evening (18:00, Hotel Milser, see below) and at the conference location on Monday morning at 8:00 to 10:00. Please contact the organizers in case you plan to arrive later.

Welcome get-together

On Sunday, Sept. 25 an informal get together will be offered in Hotel Milser starting from 18:00. A small snack will be provided.

Posters

Posters (format A0 portrait) will be up throughout Monday and Tuesday in the auditorium in order to enable for in-depth discussions. Please be sure to have your material posted before the coffee break on Monday morning.

Conference dinner

The conference dinner will be held on Monday night. The bus will leave from the conference location at 18:30. After the dinner bus transfer to the conference hotels will be provided.

Lab tour

A lab tour to IVG at Duisburg Campus of University Duisburg Essen will be organized on Wednesday afternoon (17:00 – 19:00). Please sign in during registration if you are interested in joining the lab tour.

Exhibition

Please visit the exhibition of our Sponsors LaVision, Radiant Dyes, Innolas and Micos

Sponsors

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Program

Sunday, Sept. 25: Conference

18:00 **Informal get together (Hotel Milser)**

Monday, Sept. 26: Conference

8:30 **Welcome**

Experimental investigation and modelling of the dependencies of LII signal experimental conditions: Heat conduction / accommodation coefficients

9:00 **Heat conduction issues in laser-induced incandescence**
S.-A. Kuhlmann, J. Reimann, S. Will
Technische Thermodynamik, Universität Bremen, Germany

Laser fluence

9:20 **A detailed experimental and theoretical comparison of spatially-resolved laser-induced incandescence signals**
H. Bladh[2], J. Delhay[1], Y. Bouvier[1], E. Therssen[1], P-E. Bengtsson[2], P. Desgroux[1]
[1]PC2A, Université des Sciences et Technologies de Lille, Lille, France
[2]Division of Combustion Physics, Lund Institute of Technology, Lund, Sweden

Particle size / agglomeration / modification

9:40 **Investigations of the mechanisms involved in LII particle detection**
H. A. Michelsen, M. Y. Gershenzon, P.O. Witze
Sandia National Laboratories, Livermore, CA, USA

10:00 **Influence of polydisperse distributions of both primary particle and aggregate sizes on soot temperature in low-fluence laser-induced incandescence**
F. Liu[1], M. Yang[1], F. A. Hill[2], G. J. Smallwood[1], D. R. Snelling[1]
[1]ICPET, National Research Council Canada, Ottawa, Ontario, Canada
[2] Dept. of Systems Design Engineering, University of Waterloo, Waterloo, Canada

10:20 **2-Color LII measurements of carbon black: Interpretation for quantitative measurement of fineness**
B.J. Stagg
Columbian Chemicals Company, Marietta, GA, USA

10:40 **Coffee break and poster display**

Refractive index

11:25 **Wavelength-dependence of refractive index function of soot particle by two-color laser induced incandescence**
Y. Bouvier, E. Therssen, P. Desgroux
Lab. Physicochimie des Processus de Combustion et de l'Atmosphère (PC2A), Université des Sciences et Technologies de Lille. 59655 Villeneuve d'Ascq Cedex-France

Calibration

11:45 **An LII technique independent of *ex-situ calibration* by detecting absolute light intensity**
D. R. Snelling[1], G. J. Smallwood[1], F. Liu[1], Ö. L. Gülder[2], W. D. Bachalo[3]
[1]ICPET, National Research Council Canada, Ottawa, Ontario, Canada
[2]University of Toronto, Institute for Aerospace Studies,Toronto, Ontario, Canada
[3]Artium Technologies, Sunnyvale, CA, USA

Bath gas

12:05 **Laser-induced processes in carbon generated in an argon arc**
J.D. Black[1], M.P. Johnson[2]
[1]Strategic Research Centre, Rolls-Royce plc, Derby , DE24 8BJ, UK
[2] JJ Thomson Physical Laboratory, University of Reading, Whiteknights, UK

12:25 **Lunch**

Pressure

13:40 **An investigation of soot nanoparticulate in a vacuum**
V. Beyer, D.A. Greenhalgh
School of Engineering, Cranfield University, Cranfield, UK

14:00 **Laser-induced incandescence measurements in a laminar co-annular non-premixed methane/air flame at pressures of 0.5 to 4.0 MPa**
K. A. Thomson, D. R. Snelling, G. J. Smallwood, F. Liu
ICPET, National Research Council Canada, Ottawa, Ontario, Canada

14:20 **Laser-induced incandescence and shifted vibrational CARS in laminar premixed flames at atmospheric and elevated pressures**
K.P. Geigle[1], M.S. Tsurikov[1], W. Meier[1], V. Krüger[2], R. Hadef[3]
[1]Institute of Combustion Technology, DLR, Stuttgart, Germany
[2]Robert Bosch GmbH, Gerlingen, Germany
[3]Institut de Génie Mécanique, Université Larbi Ben M'Hidi, Oum El Bouaghi, Algeria

14:40 **Laser-induced incandescence and multi-line NO thermometry for soot diagnostics at high pressures**
M. Hofmann[1], H. Kronemayer[1], B. F. Kock[2], C. Schulz[2]
[1]PCI, Universität Heidelberg, Germany
[2]IVG, Universität Duisburg-Essen, Germany

15:00 **Soot particulate size measurements in a heavy duty Diesel engine**
B. Bougie, L.C. Ganippa, A.P. van Vliet, N.J. Dam, W.L. Meerts, J.J. ter Meulen
Applied Molecular Physics, Radboud University Nijmegen, The Netherlands

15:20 **Modeling of time-resolved laser-induced incandescence (TIRE-LII) transients for particle sizing in high-pressure spray combustion environments**
T. Dreier[1], B. Bougie[2], L. Ganippa[2], N. Dam[2], T. Gerber[1], J.J. ter Meulen[2]
[1]Dept. of General Energy, Paul Scherrer Institut, Villigen PSI, Switzerland
[2] Inst. for Molecules and Materials, Radboud University, Nijmegen, The Netherlands

15:40 **Coffee break and poster display**

Non-soot LII

16:20 **Application of TR-LII for the study of carbon vapor condensation at room temperature**
A. Eremin[1], E. Gurentsov[1], M. Hofmann[2], C. Schulz[3]
[1]Institute for High Energy Density RAS, Moscow, Russia
[2]PCI, Universität Heidelberg, Heidelberg, Germany
[3]IVG, Universität Duisburg-Essen, Duisburg, Germany

16:40 **Planar laser-induced incandescence of iron particles in welding fumes**
O. Lucas[2], Z. Alwahabi[1], V. Linton [2]
Schools of Chemical [1] and Mechanical[2] Engineering, University of Adelaide, Australia

17:00 **Time-resolved laser-induced-incandescence (TR-LII) for iron-particle sizing**
B. Kock, J. Knipping, H.R. Orthner, C. Kayan, C. Schulz, P. Roth
IVG, Universität Duisburg-Essen, Germany

17:20 **Laser-induced incandescence of free and surface-adsorbed particles**
T. Schittkowski, D. Böker, D. Brüggemann
LTTT, Universität Bayreuth, Germany

Applications of LII

17:40 **In-situ determination of gas-to-particle reaction generated nanoscaled particles**
M. Charwath, T. Lehre, R. Suntz, H. Bockhorn
Institut für Chemische Technik und Polymerchemie, Universität Karlsruhe, Germany

18:00 **Two-dimensional imaging of soot volume fraction and OH in turbulent jet diffusion flames spanning low to high mixing rates**
N. H. Qamar[1], Z.T. Alwahabi[1], G. J. Nathan[2], K. D. King[1]
Schools of Chemical[1] and Mech.[2] Engineering, The University of Adelaide, Australia

18:30 **Bus transfer to Conference Dinner (return by bus at the hotels at ~ 22:30)**

Posters (Monday and Tuesday, all day)

P01 **Peak soot temperature in laser-induced incandescence measurements.**
S. De Iuliis, F. Cignoli, G. Zizak
CNR-IENI, Istituto per l'Energetica e le Interfasi, Milano, Italy

P02 **Soot volume fractions and primary particle size estimations by means of simultaneous time-resolved and 2D laser-induced incandescence**
A. Boiarciuc, F. Foucher, C. Mounaïm-Rousselle,
Depart. of Mech. and Energetics, Polytechnic School of Orléans University, France

P03 **Time-resolved laser-induced incandescence applied to in-cylinder Diesel particle sizing**
B. F. Kock, C. Schulz, P. Roth
IVG, Universität Duisburg-Essen, Germany

P04 **Gas-phase temperature imaging in sooting flames by multi-line NO-LIF thermometry**
H. Kronemayer[1], M. Hofmann[1], K. Omerbegovic[2], C. Schulz[2]
[1]PCI, Universität Heidelberg, Germany
[2]IVG, Universität Duisburg-Essen, Germany

P05 **A critical evaluation of the thermal accommodation coefficient of soot determined by the laser-induced incandescence technique**
F. Liu, D. R. Snelling, G. J. Smallwood
Institute for Chemical Process and Environmental Technology, NRC, Ottawa, Canada

Tuesday, Sept. 27: Workshop

8:30 Modeling of the LII Process
Hope Michelsen, CRF, Sandia Natl. Labs, Livermore, CA, US
Boris Kock, IVG Universität Duisburg-Essen, Germany

I.a Structure of model approaches. A Review
- which different levels of complexity are applied by the different models?

I.b Energy- and mass balance during particle heating and cooling
  - separation of heating and cooling or simulation of the complete process
- considered heat fluxes
  - temperature dependence of the heat capacity
  - solution of the differential equation-system (heat- and mass balance)

I.c Different models for heat fluxes
  - different Knudsen regions

I.d Simulation of radiation signals
  - influence of particle size distribution
  - consideration of the radial and temporal distribution of laser fluence

I.e Material properties
  - influence on the model

10:15 Coffee break and poster display

11:00 Experimental
Greg Smallwood, National Research Council, Canada
Max Hofmann, Universität Heidelberg, Germany
Coralie Shoemaecker, Université de Lille, France

II.a Excitation
- wavelength
- influence of radial and temporal energy distribution
- devices for homogenisation of laser fluence

II.b Signal Detection
- one- or two-dimensional signal detection
- detection wavelengths
- potential interferences

II.c Influence of laser fluence
- on signals: excitation curve
- on particles: change of aggregate structure and particle morphology

II.d Two-color applications for direct temperature measurement

12:30 Lunch

14:30 Signal evaluation
Stefan Will, Universität Bremen, Germany
Rainer Suntz, Universität Karlsruhe, Germany
Bas Bougie, Radboud University Nijmegen, Netherlands

III.a Determination of particle size or size distribution from measured signals
- Fitting of measured curves or
  numerical solution of Fredholm integral equation

III.b Influence of the particle shape / degree of agglomeration

III.c Determination of particle volume fraction from the measured signals
  - techniques available for the calibration
- problem of calibration (optical properties, effect of fuel and flame conditions)
- correction of temperature difference between calibration and measurement

III.d Complementary techniques

16:15 Coffee break and poster display

17:00 Comparison of different approaches of all involved group
Greg Smallwood, NRC, Canada
Christof Schulz, IVG, Universität Duisburg-Essen, Germany

IV.a Comparison of the measurements and results at the model flame
IV.b Comparison of the evaluation of the model signals

Wednesday, Sept. 27: Workshop

8:30 In-depth discussion in split groups

12:30 Lunch

14:00 Report of the results of the in-depth discussion

15:00 Definition of hot topics for upcoming meeting

16:00 Coffee break

17:00 Lab tour

Heat conduction issues in laser-induced incandescence

S.-A. Kuhlmann, J. Reimann, S. Will*
Technische Thermodynamik
Universität Bremen, Germany

Uncertainties in modelling heat conduction in connection with the application of laser-induced incandescence (LII) to primary particle sizing are discussed. Besides a comparison between commonly used models an experimental study employing specified carbon blacks was performed. An overall good correlation of LII-values and those from electron microscopy has been found. Based on these data an effective accommodation coefficient of $\alpha = 0.23$ is determined and the influence of aggregate size on primary particle sizing has been quantified.

Introduction

In the time interval evaluated for primary particle sizing by laser-induced incandescence (LII) heat conduction is the dominant cooling mechanism. Thus uncertainties in modelling this process may result in considerable errors in the sizes determined. It is the aim of the present paper to critically assess two models on heat conduction widely used and to experimentally provide quantitative information both on the thermal accommodation coefficient α and on the influence of aggregate size on primary particle sizing.

Numerical

Modelling the heat conduction between primary particles and the surrounding gas, different approaches are compared, namely the most widely used formula proposed by McCoy and Cha [1] and the approach developed by Fuchs [2]. Based on numerical implementations it is shown that considerable differences may occur depending on relevant temperatures and particle sizes. The models may be reconciled, however, with proper choices of "effective" thermal accommodation coefficients.

Experimental

Systematic measurements on re-dispersed carbon blacks with specified primary particle sizes were carried out. To that end a carbon black aerosol was produced by dispersing powders, spraying the dispersion into a carrier gas by means of an aerosol generator and flowing it through a heating section. Pointwise experiments were performed using a Nd:YAG laser at 532 nm and a standard detection scheme with a photomultiplier.

Results

When LII results, employing a typical primary particle size distribution, are related to the carbon black manufacturer's data determined by transmission electron microscopy (TEM) an effective accommodation coefficient can be obtained. Using a value of $\alpha = 0.23$ (based on the model of McCoy and Cha) for the accommodation coefficient delivers an overall good correlation.

Generally, aggregate size is expected to influence heat transfer from primary particles, mainly due to shielding effects. Numerical analyses on this issue were recently performed by Filippov et al. [3] and Liu et al. [4]. In order to experimentally address this question residual deviations in the above correlation of LII- and TEM-results were linked to aggregate sizes determined by two independent methods. Thus it could be shown that these deviations in LII-values are directly correlated to the number of primary particles per aggregate. These results may contribute to check the numerical models developed and to provide a guideline for the inclusion of aggregate effects in practical applications of LII for primary particle sizing.

Partial financial support by the German Research Foundation is gratefully acknowledged.

References

1 B.J. McCoy C.Y. Cha, Chem. Eng. Sci. 29, 381-388 (1974)
2 N.A. Fuchs, Geophys. Pura Appl. 56, 185-193 (1963).
3 A.V. Filippov, M. Zurita, D.E. Rosner, J.Colloid Interface Sci. 229, 261-273 (2000).
4 F. Liu, G.J. Smallwood, D.R. Snelling, J. Quant. Spec. & Rad. Trans. 93, 301-312 (2005).

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A detailed experimental and theoretical comparison of spatially-resolved laser-induced incandescence signals

H. Bladh[2*], J. Delhay[1], Y. Bouvier[1], E. Therssen[1], P-E. Bengtsson[2], P. Desgroux[1]
[1]PC2A, UMR CNRS 8522, Université des Sciences et Technologies de Lille, Lille, France
[2]Division of Combustion Physics, Lund Institute of Technology, Lund, Sweden

To improve LII as a quantitative tool for extraction of soot properties, the models describing the heat and mass transfer between the particles and the surrounding gas must be tested against well-characterized experiments. In this work we have made a detailed investigation in which experimental results have been compared with simulated ones in two different configurations; backward-LII and 90º-LII. Although the results show a qualitatively good agreement, it is realized that an improved physical description is needed for the LII signal behavior.

Introduction

The laser-induced incandescence (LII) technique has evolved as a powerful technique for quantitative measurements of soot volume fraction as well as of particle size. Still, however, fundamental knowledge of the processes underlying the signal behavior is partly unknown, which motivated the present work.

Experimental and theoretical approach

Experiments were performed in an axisymmetric methane diffusion flame. The central part of a Nd:YAG laser beam at 1064 nm was directed through the flame, and the spatial distribution of the laser beam was monitored. The spatially resolved LII distributions were imaged in two directions of observation, see Fig.1.

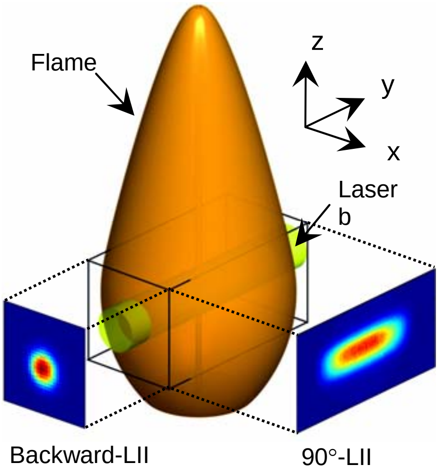

Fig. 1: Backward-LII and 90º-LII and their relation to the measurement volume.

Two-dimensional images of spatial LII distributions were recorded for both experimental configurations as a function of laser pulse energy and using different gate timings.

The heat and mass transfer model for the soot particles heated by laser radiation calculates time-resolved LII signals for a given set of input parameters [1]. Integrated LII signal intensities were calculated from simulated measurement volumes created using data from the experiments, such as the spatial distribution of laser energy.

Results

An example of experimental data is shown in Fig. 2, where simultaneous images are shown for both backward-LII and 90º-LII. In both images strong vaporization can be seen at the center of the spatially resolved LII signals.

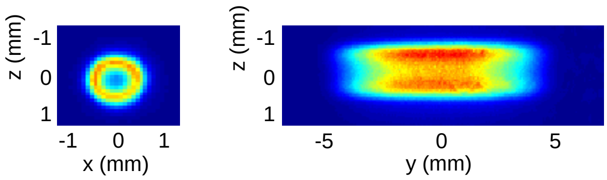

Fig. 2: Spatially-resolved LII signals from experiments using backward-LII (left) and 90º-LII (right).

In the comparison of experimental data with simulated ones, it was shown that the effects of the non-homogeneous laser energy distribution and the vaporization characteristics were qualitatively well reproduced by the model. The presented results are important in the further development of the LII model.

Acknowledgments

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1 H. Bladh, P.-E. Bengtsson, Appl. Phys. B 78, 241-248 (2004).

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Investigations of the mechanisms involved in LII particle detection

H.A. Michelsen*, M.Y. Gershenzon, P.O. Witze
Sandia National Laboratories, Livermore, CA 94551

We have made measurements of the temporal response of Laser-Induced Incandescence to pulsed excitation of soot in a flame and developed a corresponding LII model that accounts for particle heating by laser absorption, oxidation, and annealing and cooling by sublimation, radiation, and conduction to the surrounding atmosphere. The model also includes mass loss by oxidation, sublimation, and nonthermal photodesorption of carbon clusters. The results of this study allow us to identify the largest uncertainties associated with the understanding of LII and predict the influence of measurement parameters on LII signal under varying conditions.

Experimental Studies

To test current models and investigate the influence of experimental conditions on LII behavior, we have measured time-resolved LII signals from soot in a nonsmoking coflow ethylene diffusion flame over a wide range of laser fluences as high as 4 J/cm^2 [1]. A Nd:YAG laser was injection seeded to provide a smooth laser temporal profile with a pulse duration of 7 ns, the output was doubled to generate 532-nm light, and the beam was passed through an aperture and relay-imaged into the flame to produce a smooth laser spatial profile. LII temporal profiles were recorded with a fast photodiode with adequate temporal resolution to capture signal evolution during the laser pulse. We used these results to aid in the development of a model that predicts the temporal behavior of LII from soot on a nanosecond time scale. The model accounts for particle heating by laser absorption, oxidation, and annealing and cooling by sublimation, conduction, and radiation. The model also includes mechanisms for convective heat and mass transfer, melting, and nonthermal photodesorption of carbon clusters [2].

Another set of experiments was performed to investigate the fast photodesorption mechanism in more detail. In these experiments the particles were heated with 532-nm pulses of ~70 ps duration from a regeneratively amplified modelocked Nd:YAG laser over a range of fluences as high as 0.6 J/cm^2. The signal was collected with a streak camera with a temporal resolution of ~15 ps.

Model Development

Models typically used to describe LII are based on a model initially developed by Melton[3] in which energy- and mass-balance equations are solved to account for particle heating by laser absorption and cooling by conduction to the surrounding atmosphere, radiative emission, and sublimation. Particle size reduction during sublimation is also calculated. LII signal is derived from calculated temperatures and sizes using the Planck function weighted by the emissivity and the detector's wavelength response. The Melton model [3] uses (1) temperature-independent values for density and specific heat to determine the internal energy of the particle, (2) a Rayleigh approximation for laser absorption and radiative emission rates, (3) the approximation of kinetic control of C_3 only from the surface for sublimation rates, (4) a thermal accommodation coefficient and heat capacity associated with temperatures near room temperature, and (5) a formulation for conductive cooling appropriate for a transition regime between Knudsen and continuum flow.

We have developed a model [1,2] that similarly solves the energy- and mass-balance equations but also includes (1) temperature-dependent thermodynamic parameters for calculating sublimation, conduction, and internal energy storage by the particle, (2) wavelength-dependent optical parameters to describe absorption and emission of radiation based on a Rayleigh-Debye-Gans approximation to account for aggregation, (3) convective heat and mass flow (Stefan flow) during the sublimation of multiple cluster species (C, C_2, C_3, C_4, and C_5) from the surface, (4) a thermal accommodation coefficient appropriate for high temperature conductive cooling, (5) a conductive cooling mechanism assuming free molecular flow at low pressure and a transition regime at high pressure, (6) nonthermal photodesorption resulting in loss of heat and mass by carbon clusters leaving the particle, (7) phase changes (i.e., annealing and melting) and their effects on absorption, radiation, sublimation, and photodesorption, and (8) oxidative heating at the particle surface.

References

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Influence of polydisperse distributions of both primary particle and aggregate sizes on soot temperature in low-fluence laser-induced incandescence

F. Liu[1], M. Yang[1], F. A. Hill[2], G. J. Smallwood[1*], and D. R. Snelling[1]
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[2] Department of Systems Design Engineering, University of Waterloo, Waterloo, Ontario, Canada

An improved aggregate-based low-fluence laser-induced incandescence (LII) model was developed. The *shielding effect* on the heat conduction between aggregated soot particles and the surrounding gas was modeled using the concept of the equivalent heat transfer sphere. The diameter of such equivalent sphere was determined from detailed direct Monte Carlo simulation in the free molecular regime as functions of the aggregate size and the thermal accommodation coefficient (TAC) of soot. The effect of primary particle diameter polydispersity is in general important and should be considered. The effect of aggregate size polydispersity is relatively unimportant when the heat conduction between the primary particles and the surrounding gas takes place in the free-molecular regime; however, it becomes important when the heat conduction process occurs in the transition regime. Application of the model was also applied to re-determine of the TAC of soot in an atmospheric pressure laminar ethylene diffusion flame.

Introduction

It is shown by thermophoretic sampling/transmission electron microscopy techniques that flame-generated soot consists of polydispersed primary particles and aggregates. There is currently lack of a quantitative understanding of the shielding effect on heat conduction between aggregated soot particles and the surrounding gas. This study made an attempt to develop an aggregate based low-fluence LII model to investigate the shielding effect and examine the implication of such effect to LII based particle sizing techniques.

LII Model

The low-fluence LII model formulated for a soot aggregate was developed by introducing the concept of heat conduction equivalent sphere. The diameter of such sphere was determined from detailed direct Monte Carlo simulation in the free molecular regime as functions of the aggregate size and the TAC of soot. Heat conduction loss rate from the soot aggregate to the surrounding gas was calculated using the Fuch's approach. The distributions of the primary particle diameter and the aggregate size were assumed log-normal.

Results

The relative importance of aggregate size (N) and primary particle diameter (d) distribution under conditions of a typical flame, where heat condition occurs in the free-molecular regime, is shown in Fig. 1. It shows that the polydispersity of aggregate size is relatively unimportant while the polydispersity of primary particle diameter is more important.

Fig. 2 shows the relative importance of the distribution of N and d under the condition of a low gas temperature where the heat conduction takes place in the transition regime. While the polydispersity of d is still more important that that of N, the effect of polydispersity of N becomes more pronounced compared to the results shown in Fig.1. The results shown in Fig. 2 implies that the LII based nanoparticle sizing techniques for primary particle diameter measurement suffers ambiguity when applied to situations where the polydispersity of N is significant.

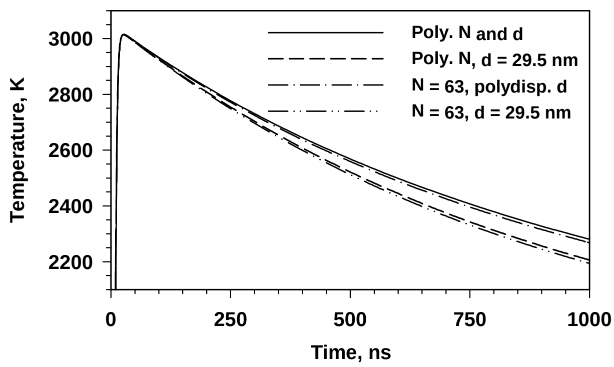

Fig. 1 Relative importance of N and d distributions in the flame case: T_g = 1720 K.

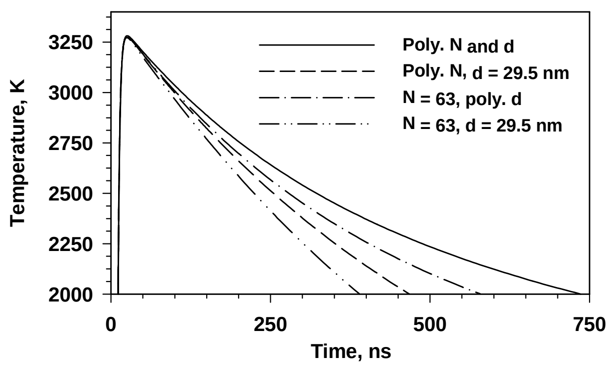

Fig. 2 Relative importance of N and d distributions in the low gas temperature case: T_g = 440 K.

Application of this aggregate based low-fluence LII model to analyze our data found that the TAC of soot to be 0.38.

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2-Color LII measurements of carbon black: Interpretation for quantitative measurement of fineness

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The 2-color LII technique is utilized to provide on-line measurement of the fineness level of carbon black during its manufacture. The BET surface area does not follow the expected "*1/d*" behavior, and the soot volume fraction measured by LII has been found to be dependent upon laser fluence. Reasons for these anomalous behaviors are not yet known.

Introduction

The specific surface area of aggregated nanoparticles, related to primary particle diameter, is a key parameter in the production of carbon black. Currently, there is no available technique to provide on-line, real-time measurement of this parameter during the production of carbon black. Rather, periodic sampling, followed by adsorption measurements (such as the BET method), is typically utilized by the carbon black industry to provide this information. It is well known that 2-color LII has been utilized to measure soot volume fraction (svf) and primary particle size of an aerosol cloud (see e.g. [1-2]).

Experimental Equipment

A small amount of carbon black aerosol is continuously drawn from a commercial carbon black reactor system using a venturi eductor sampling device. A critical orifice, attached to the inlet of the venturi eductor, controls the flow rate of aerosol that is sampled. Because of the high dilution ratio utilized (approximately 200:1), the dilution gas comprises >99% of the background gas during the LII measurements. The gas and aerosol stream are cooled to ambient temperature prior to entering the LII instrument.

A commercial, 2-color LII instrument (Artium Technologies) is utilized to make measurements at a pulse repetition frequency of 20 Hz. For each laser pulse, the incandescence signals are measured at wavelengths of 400 nm and 780 nm. The incandescence signals, measured over a duration of approximately 1000 ns at intervals of 10 ns, are converted to intensity values using previously determined calibration factors. From the intensity values, the soot volume fraction and the particle temperature are calculated. The decay rate of the particle temperature is utilized as a measure of particle fineness [1].

Measurements

The LII measurements were performed during the production of two different grades of carbon black (N234 and N299). During the production process, the specific surface area of the carbon black was purposely varied a small amount from the target to determine the capability of the LII technique. For comparison, the BET measurements of specific surface area were performed on carbon black samples collected during the tests. Although the 2-color LII technique is able to resolve small changes in specific surface area, BET surface area does not exhibit the expected "*1/d*" dependence. Furthermore, the power of the exponent seems to be different for different grades of carbon black.

One of the benefits of using 2-color LII (as opposed to a single detector technique) is the ability to utilize low laser fluence levels, where carbon sublimation is negligible [3]. In this regime, the soot volume determined by LII should not vary with laser fluence. As seen in Figure 1, the indicated svf shows a marked and unexpected dependence upon laser fluence.

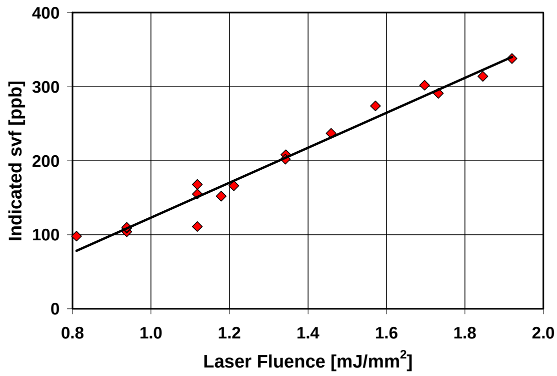

Fig. 1: Indicated Soot Volume Fraction

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Wavelength-dependence of refractive index function of soot particle by two-color laser-induced incandescence

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A new method is proposed to obtain the variation of the refractive index function of soot particles *E*(*m*) with wavelength. The method consists to select laser energies at different wavelengths ensuring the equality of the LII signals in the low fluence regime. Such equality is consistent with the fact that the soot particle has reached the same temperature independent on the laser wavelength, e.g. the soot particle has absorbed the same energy. The measurement of the laser energies insuring a perfect concordance of the LII intensities allows the determination of the corresponding variation of *E(m)* with wavelength. The method is applied in a methane diffusion flame by using laser radiation at 532 and 1064 nm.

Introduction

A few papers report on the wavelength-dependence of *E(m)*. It has been shown that results present large discrepancies [1-3]. Particularly accurate knowledge of *E(m)* at 1064 nm is necessary since this wavelength is the most recommended to prevent background fluorescence to be collected with the LII signal. In this work, the recently proposed two-color LII method [4] is extended to derive data on soot optical properties. It is shown that by using two different laser wavelengths (532 and 1064 nm), the ratio of the soot absorption functions *E*(*m*,532) / *E*(*m*,1064) can be locally determined in any flame. This new method is applied in a methane diffusion flame presenting an axisymmetric soot distribution.

Method

The original method proposed aims to obtain the variation of *E(m)* with wavelength and is based on the LII signal variation with laser excitation wavelength. For the demonstration the two laser wavelengths used in this work were 532 and 1064 nm. But the method can be extended to any couple of wavelengths. The incandescence radiation is collected with the same spectral efficiency independent on the laser wavelength (same $\lambda_{detection}$).

The power absorbed by the particle (radius r_p) is related to the laser irradiance q (Wm^{-2}), and to $E(m,\lambda_{laser})$ according to:

$$H_A = Q_{abs}\,\pi\, r_p^2\, q \quad \text{with} \quad Q_{abs} = 4 \cdot E(m,\lambda_{Laser}) \cdot \frac{2.\pi.r_p}{\lambda_{laser}}$$

where Q_{abs} is the soot absorption efficiency.

Thus, for both selected laser wavelengths we ensure: (1) absence of particle sublimation (2) coincidence of temporal LII signals, (3) same spatial laser energy distribution and (4) the same intensity and volume emission of the two thermal radiations, the ratio of the *E(m)* functions at any laser wavelength is given by the inverse ratio of the laser energies (selected to ensure conditions 2 and 4) times the ratio of the wavelengths.

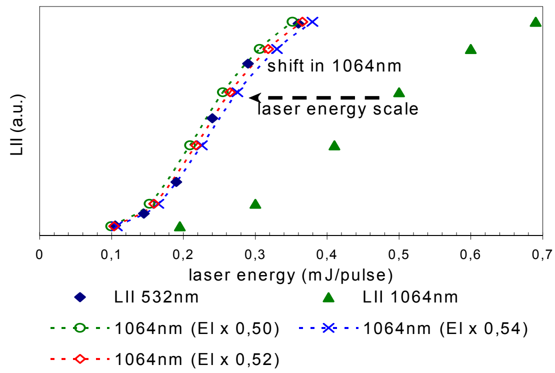

Fig. 1. Determination of the E(m) ratio.

The fluence curves have been thoroughly obtained in the low fluence range. Prompt-LII intensity is plotted in Fig.1 as function of the laser energy/pulse. It can be seen that the fluence curve at 1064 nm perfectly fits that one at 532 nm by reporting it as function of a new abscissa namely (0.52 x E_{laser}(532)) (red curve). Thus, LII signals collected successively upon 532 or 1064 nm irradiance are identical for a ratio of the laser energies E_{laser}(532)/ E_{laser}(1064) = 0.52 leading to a ratio of *E*(*m*,1064)/ *E*(*m*,532) = 1.04. Without any normalization, LII temporal profiles, measured for any couple of laser energies insuring the equality of the peak LII intensities, were found to be identical.

The method can be applied locally and will be extended to the study of *E(m)* variation in different flames (equivalence ratio, composition…).

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An LII technique independent of *ex-situ calibration* by detecting absolute light intensity

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This paper presents a novel LII technique for the determination of soot volume fraction by measuring the absolute incandescence intensity, avoiding the need for *ex situ* calibration typically using a source of particles with known soot volume fraction. The technique developed in this study further extends the capabilities of existing LII for making practical quantitative measurements of soot. The spectral sensitivity of the detection system is determined by calibrating with an extended source of known radiance and this sensitivity is then used to interpret the measured LII signals. Although it requires knowledge of the soot temperature, either from a numerical model of soot particle heating or experimentally determined by detecting LII signals at two different wavelengths, this technique offers a calibration independent procedure for measuring soot volume fraction. Application of this technique to soot concentration measurements is demonstrated in a laminar diffusion flame.

Introduction

The LII technique was traditionally calibrated using a particle source of known concentration. For this conventional LII method, as long as there are no significant changes between the conditions of calibration and the LII measurements, the measured volume concentration should be relatively accurate. However, all particulates are not created equal. Calibrating in a flame at high temperature and then measuring at lower temperatures can introduce inaccuracies due to different peak temperatures. Furthermore, variation in the ambient pressure can also have an effect, as it impacts upon the sublimation temperature of elemental carbon. Because of these limitations of the conventional LII method, caution must be taken when applying the calibration constant obtained under a condition substantially different from the actual measurement condition in terms of the local gas temperature, the chemical compositions of the PM, and the ambient pressure. The object of this study is to develop a calibration-independent LII technique.

Methodology

The fundamental idea behind the present LII technique is that the soot volume fraction can be determined if the absolute spectral intensity of the incandescence signal emitted from the laser heated soot particles is measured and the temperature of the soot particles is known. To measure the absolute LII intensity the detection system must be calibrated using a radiation source of known radiance. The soot particle temperature is required to calculate the theoretical spectral emission intensity per unit volume of soot. The temperature of soot particles can be measured by detecting the LII signals at two different wavelengths. The detection system is calibrated using a strip filament lamp of known brightness temperature and thus known radiance. The optical set-up for absolute light intensity calibration is shown schematically in Fig. 1.

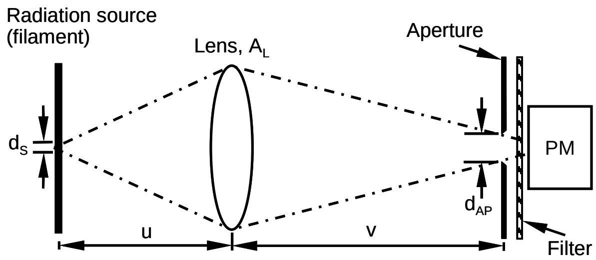

Fig. 1 Schematic of the optical setup for the calibration of absolute light intensity.

Results

Results of soot volume fraction measurement in a laminar diffusion flame using this LII technique are shown in Fig. 2. The LII results are in good agreement with that measured by 2D attenuation (4 ppm).

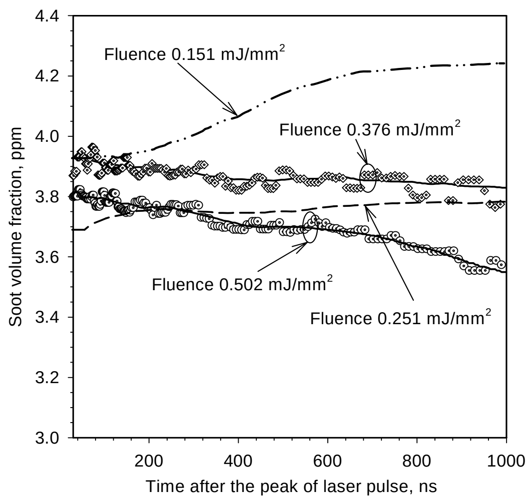

Fig. 2 Variation of the soot volume fraction measured using the present LII technique with laser fluences.

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Laser-induced processes in carbon generated in an argon arc

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The interaction of pulsed Nd:YAG laser radiation with particles produced by a commercial carbon aerosol generator employing a discharge between graphite electrodes in argon has been studied under a variety of conditions. Other processes besides LII are observed and response to the laser pulses is very dependent on the composition of the carrier gas surrounding the particles.

Introduction

The Palas GfG1000 commercial 'graphite' aerosol generator is a convenient source of small carbonaceous particles. This device forms particles in an electrical discharge between graphite electrodes in a flow of argon. Air can be added to the gas flow after the discharge region to dilute the particle concentration. This device is a convenient aid for setting up LII systems for in-situ measurement in, for example, aero-engine exhausts, but the nature of the particles is different to combustion soot.

An unfocused Nd:YAG laser operating at 1064 nm was used to generate LII at the outlet of an alumina tube which carried the exhaust from the GfG 1000 through a tube furnace so that the temperature of the carrier gas could be varied. Fig.1 (taken with a Sony Mavica digital camera) shows visible LII at the outlet of the heated furnace. The 7-mm diameter laser beam traverses the lower right hand corner of the picture diagonally from right to left. LII appears as a bluish cylinder.

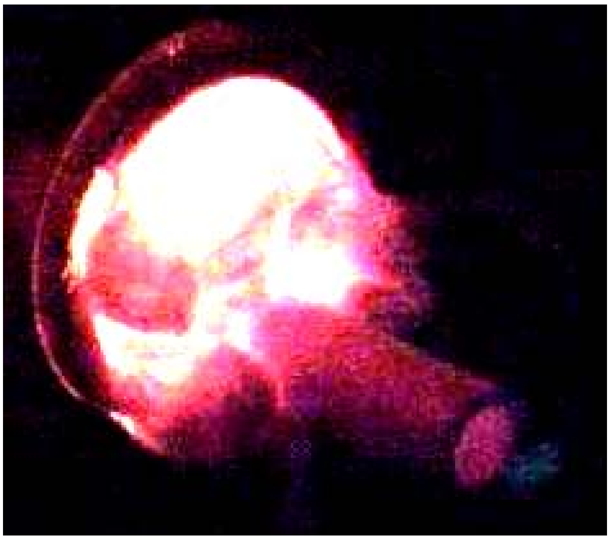

Fig. 1: LII from particles in an argon stream emerging from the tube in the lower right corner. The bright part of the image is insulation at the mouth of a tube furnace at ~1000°C, giving a 'colour temperature' reference to the LII, which appears much bluer.

Discussion

In a previous publication [1] we showed spectra of the LII in pure argon and 1:1 argon/air. With pure argon the spectrum peaks around 560 nm with some broad structure around the peak and a tail extending into the UV. When air is added LII signal levels drop drastically, as does the emission spectrum at wavelengths shorter than 550 nm. After initial dilution, LII scales linearly with the relative air flow.

This suggests that air acts to quench a laser-induced effect which dominates in a pure argon flow.

Fluence dependence has been studied in the range 0.03 – 0.3 J cm^{-2} for both argon and argon/air mixtures. In images of the LII taken in the first microsecond following the laser pulse very bright spots with dimensions < 0.1 mm are observed even at lower fluence levels in both argon and argon/air, although to a lesser extent when air is present.

The GfG 1000 probably produces a variety of different forms of carbon, e.g. graphenes, nanotubes, etc. Electron micrographs show very small particles ~10 nm. Investigations are continuing.

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Laser-induced incandescence (LII) measurements in high vacuum conditions

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We have undertaken a preliminary study of Laser-Induced Incandescence (LII) using carbon black in high vacuum (circa 5×10^{-4} mbar). Our aim was to understand the temperature behaviour of carbon nanoparticles under conditions dominated by radiative cooling. We have found that under these conditions the signal from LII is markedly enhanced (over 2,000 times) and that cooling behaviour can be measured into the 100 µs timescale.

Introduction

LII so far has been studied, modeled and well characterized in many standard conditions: flames, vehicle exhaust conditions, for high pressures and high temperatures domain etc. However the present study describes a novel LII application under high vacuum after the evidence that in 1988 Rohlfing [1] observed carbon clusters enduring LII from ~3900 K to ~2700 K over 42 µs during a study of laser vaporization of graphite. The application of such technique for particulate matter measurements, namely called LII under vacuum, would be solely as an extractive sampling to an external evacuated optical cell.

Theory

The aim prior to the experiment taking place was to deliberately stop conductive heat losses occurring and hence retard the light signal from typical 50 – 200 ns scale to a much longer 40 – 100 µs duration. Thus the main energy loss mechanism should become long-lived blackbody radiative cooling. This approach has the added benefit of significantly increasing the LII signal. Calculated gains in LII signals for a scan duration of 100 us and an input fluence of 0.2 J/cm^2 range are from over 10 000 for a primary particulate radius below 20 nm and ~2000 for a primary particulate radius approaching 100 nm. This preliminary study took place circa 5×10^{-4} mbar, where modeling demonstrated that conductive heat losses are negligible for primary particulates smaller than 10 nm in diameter for temperatures above 2000 K.

Experimental setup

Obtaining a suspension of particulates in a vacuum is clearly a demanding task. One approach is to use a particle beam akin to a molecular beam. This is complex and expensive to implement. Thus to commence this study we choose a simpler approach based on low cost components. Our experiments were conducted in a rotating cylindrical glass bottle. The glass bottle was filled with a few grams of carbon black, evacuated to nearly 10^{-4} mbar and then hermetically sealed. Centrifugal and frictional forces created by the rotation lifts the carbon black aggregates to the top of the bottle, allowing them to drop into the laser beam and initiate LII. The incandescence signals were analyzed using a two-color transient pyrometric system and an intensified camera (see fig.1).

Results and observations

Large carbonaceous aggregates could be isolated and analyzed, these aggregates exhibited a behavior close to a radiative cooling and presented some novel behaviours which may indicated morphology changes to the carbon. Experimental evidence also suggests that light shielding within an aggregate occurs and plays a strong role for the strongly aggregated carbon black.

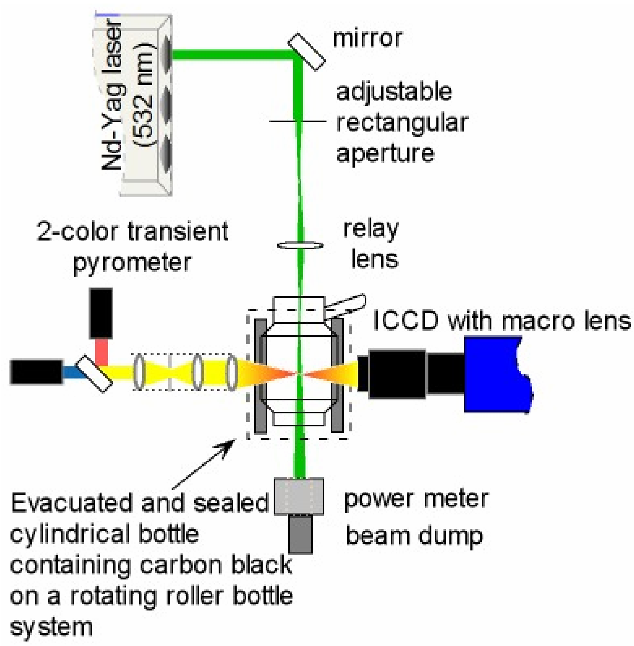

Fig. 1: Experimental setup

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Laser-induced incandescence measurements in a laminar co-annular non-premixed methane/air flame at a pressure of 0.5 - 4.0 MPa

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Auto-compensating laser-induced incandescence (AC-LII) was applied to a high pressure non-premixed flame to measure soot volume fraction (SVF) and effective primary particle diameter (dp_{eff}). Measured SVF and dp_{eff} showed annular structures and good radial symmetry. LII determined SVF were lower than previous LOSA SVF measurements (60% lower at p = 0.5 MPa up to 30% lower at p = 4.0 MPa). The LII SVF curve shapes were similar to the LOSA SVF curves, though the former had lower measurement spatial resolution at the higher pressures (*i.e.* P = 2.0 and 4.0 MPa). Effective primary particle diameter increased with pressure and radius (e.g. 20 nm at r = 0 mm, p = 0.5 MPa to 100 nm at p = 4.0 MPa).

Introduction

The objective of the present work is to apply the AC-LII method [1] to the SVF and size measurement of soot in a non-premixed high pressure flame. Measurements were compared to previous line-of-sight attenuation SVF measurements (LOSA) for the same burner [2].

Experiments

A laminar co-annular non-premixed flame was operated with methane and air mass flow rates of 0.55 mg/s and 0.4 g/s, respectively, in an optically-accessible pressure vessel for pressures of 0.5, 1.0, 2.0, and 4.0 MPa. The experimental apparatus is described in [2]. Soot particles were heated by a pulsed Nd:YAG laser frequency doubled to a wavelength of 532 nm and formed into a vertical sheet. The laser intensity normal to the sheet was approximately Gaussian (σ = 36 μm). The LII emission was collected perpendicular to the laser sheet and imaged onto an optical fiber (100 μm core diameter). The fiber output was first split and filtered with interference filters having central wavelengths of 445 and 798 nm and widths of 60 nm and then imaged onto two fast photomultipliers. LII emission was sampled at 1 ns intervals. The detection system was calibrated for absolute emission intensity with a calibrated integrating sphere light source located coincident with the flame centerline. Measurement scans were made at a height of z = 6.0 mm above the burner tip along an axis through the flame centerline (*i.e.* x = 0) with measurement spacing of Δy = 50 μm. The peak laser fluence, before entering the flame, was 0.59 mJ/mm^2. The fluence dropped by as much as 20% due to attenuation in the flame. Additional data points were collected at the end of each scan with a fluence of 0.67 mJ/mm^2 to compensate for flame attenuation. The theory used in the evaluation of the LII emission is described in [1].

Results

SVF measurements for p = 1.0 MPa are included in Fig. 1 with LOSA SVF measurements at z = 5.5 and 6.0 mm. The data symmetry despite position variant fluence demonstrates that AC-LII works well in an unknown attenuating environment. Based on curve shapes (location of peak SVF and ratio of peak to centerline SVF), the LII data appears to fall between the two LOSA curves. The LII measurements are 30-40% lower in magnitude.

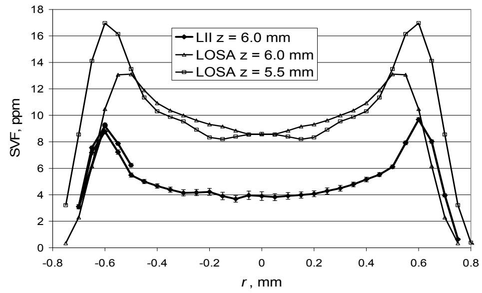

Fig. 1: LII and LOSA SVF, p = 1.0 MPa.

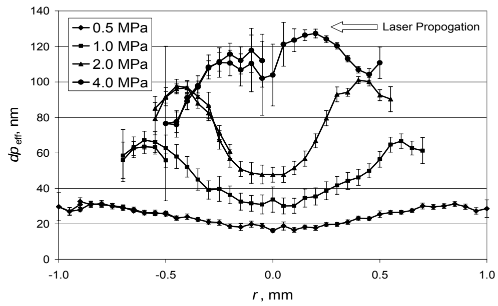

Fig. 2: Effective primary particle diameter.

Fig. 2 shows the measured dp_{eff}. Symmetry is excellent except at p = 4.0 MPa. There is a pronounced increase of dp_{eff} with increasing P and r. Overall these preliminary measurements made in a challenging high-pressure and high-property gradient environment are encouraging for future high pressure LII research.

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Laser-induced incandescence and shifted vibrational CARS in laminar premixed flames at atmospheric and elevated pressures

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An experimental investigation of various laminar premixed flames at atmospheric and elevated pressures (1-5 bar) has been conducted. Quantitative soot volume fraction measurements were obtained using laser-induced incandescence coupled with a quasi-simultaneous absorption measurement for calibration; the data were corrected for signal trapping using an "onion peeling" algorithm. Temperature measurements were obtained using shifted vibrational coherent anti-Stokes Raman scattering. Results are presented for stable homogeneous flames using air as oxidizer and ethylene, propylene, and toluene as fuels. The data identify trends and features useful for the validation of numerical models of soot formation.

Introduction

A study of laminar premixed flames under different operating conditions was performed. The study improves upon existing applied laser diagnostic techniques and enables a validation of kinetic soot models.

Experiments

Stable flames, chosen for the validation of kinetic soot models, were studied under the following conditions: six ethylene/air flames at 1, 3 and 5 bar, five propylene/air flames at the same pressures and three toluene/air flames at 1, 1.5 and 3 bar.

Laser-induced incandescence excited at 1064 nm, calibrated with an extinction measurement on the same optical pathway and at a wavelength of 532 nm, was used for the determination of soot volume fractions. Applying a refractive index of 1.60-0.59i [1] and a correction for signal trapping between the experimental plane and the detector [2], determination of the soot concentration profiles along the flame axis was possible. Since the extinction measurement was performed with a laser sheet and for each flame in situ, in principle, a calibration is possible at any height in the flame. Practically, a position between young forming soot and aged soot, thus close to the start of the soot plateau, was chosen as representative of the whole flame with one single calibration position. Clear trends of the calibration constant value with operating conditions could not be observed. Flame temperature measurements on the burner axis were performed with shifted vibrational coherent anti-Stokes Raman scattering (SV-CARS) [3].

Because the flames proved to be sufficiently stable and reproducible, each diagnostics could be applied consecutively with only a few minutes' gap in between for exchanging the respective optics. At the chosen rich equivalence ratios and flow rates the maximum temperature remained below 1730 K. Determined soot plateau concentrations varied between 0.6 ppm for the atmospheric ethylene flames and 11 ppm for the richest 5 bar ethylene flame. Clear trends could be observed with respect to the thickness of the flame front (temperature rise) in changing from the relatively simple fuel ethylene to toluene and from low to high pressure. The first variation increases the width of the flame front while the second influences it in the opposite direction. Soot concentrations typically increase with equivalence ratio (moderately), pressure (strongly) and complexity of fuel. Sample data from the study are presented in Figure 1.

In addition to the benefit for kinetic model validation, the gained accurate temperatures can be used to improve the typical uncertainty of estimated ambient temperatures for deduction of the particle size from LII decay curves in future experiments in the now defined flames.

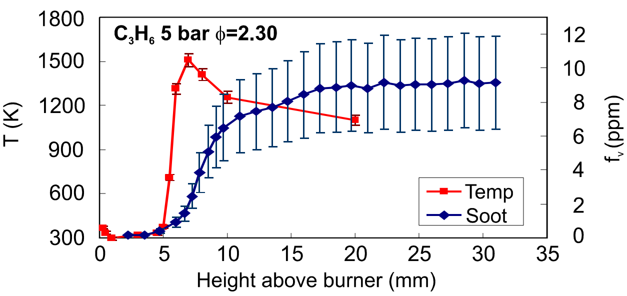

Fig. 1. Correlated soot concentration and temperature profiles along the burner axis for one studied flame.

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Laser-induced incandescence and multi-line NO LIF thermometry for soot diagnostics at high pressures

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Laser-induced incandescence (LII) is investigated in laminar premixed flat ethylene/air flames at pressures up to 5 bar in a recently constructed high-pressure burner. Time-resolved LII signals were compared with a numerical model considering the influence of pressure on the LII signal decay. Peak particle temperatures after laser heat up were measured by the detection of the LII signal at two wavelengths. Laser-induced emission spectra enabled to check for interfering signals. Gas-phase temperatures were required to compare the LII signal decay with the model and were measured using multi-line NO-LIF thermometry.

Introduction

Laser-induced incandescence (LII) has proven to be a powerful tool for soot diagnostics. It has been used successfully for measuring volume fractions of soot in flames and exhaust gases and particle sizes have been deduced from the temporal behavior of the LII signal. While applications in high-pressure environments (namely IC engines) are frequently performed, systematic investigations of LII at elevated pressures have only been performed in [1]. Important for understanding and interpreting LII signals is the temperature information of both soot particles and the surrounding gas.

Experimental

We set up a high-pressure burner with a premixed, laminar sooting ethylene/air flame. For stabilization the central sooting flame was surrounded by a premixed, non-sooting methane/air flame. Optical access was provided by four quartz windows. The flame at 1, 2 and 5 bar is shown in figure 1.

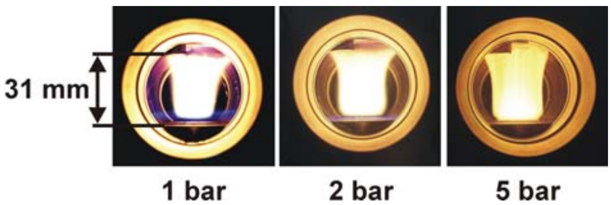

Fig. 1 . Premixed ethylene-air flame with an equivalence ratio of ϕ = 2.1 at 1, 2 and 5 bar.

LII signals were generated by a Nd:YAG laser at 1064 nm at elevated pressure up to 5 bar with wavelength, excitation-energy-density- and time-resolved detection. Time-resolved LII signals were detected at two different wavelengths with fast photomultipliers in order to obtain the peak particle temperature after laser heat up. The optical system was calibrated by a black body radiator.

Gas temperatures were measured using multi-line NO thermometry. Therefore, the sooting flame was seeded with some hundred ppm NO. The NO molecules were excited in the A-X(0,0) band by a tunable narrowband KrF*-excimer laser (248 nm) which was frequency-shifted to 225 nm in a 10-bar hydrogen Raman cell. The LIF-signal was recorded with an intensified CCD-camera. Gas temperatures were deduced by fitting simulated NO-LIF spectra to the experimental excitation spectra using the LIFSim program [2].

TR-LII measurements provide systematic information about the pressure influence on the LII technique. Excitation with different laser-energy densities gives information about soot evaporation as well as subsequent particle cooling at different pressures. Current numerical models simulating LII signals were compared with experimental data. To obtain particle sizes using the signal decay curve it is important to use the correct heat transfer model as the heat transfer mechanisms change with increasing pressure.

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Soot particulate size measurements in a heavy duty Diesel engine

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Time-resolved laser-induced incandescence (Tire-LII) was measured in an optically-accessible heavy-duty Diesel engine. The fundamental of a Nd:YAG laser was used to excite the soot particles and the particle size was determined from the temporal decay of the induced incandescence intensity as a function of crank angle. The time response of the measurement system was too slow in order to follow the decay of the LII signals. Therefore, a deconvolution algorithm of the measured Tire-LII curve was applied which compensated for the system response time.

Introduction

The purpose of this paper is to show the feasibility of performing Tire-LII measurements inside the cylinder of a heavy-duty Diesel engine at pressures varying between 1 and 70 bar. A correction for the time response of the system was made by applying a Wiener deconvolution algorithm.

Experimental setup

The experimental setup for the experiment is shown in Fig. 1. The Tire-LII measurements were performed in a heavy duty Diesel engine at two different engine loads. The first cylinder has been modified and has optical access through four quartz windows, one in the cylinder head, one in the piston and two at the side of the cylinder. A slit in the piston crown provides optical access through the side windows during the early stages of the combustion cycle.

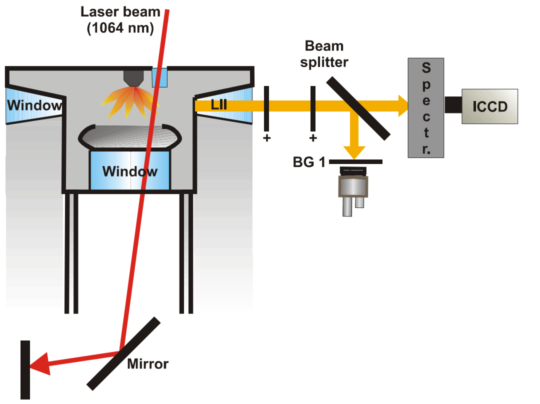

Figure 1: Experimental setup for the LII-experiments.

The soot particles were excited by the laser radiation traversing top-down through the engine and the resulting LII radiation was detected through one of the side windows. After being focused by a lens system the incandescence was simultaneously detected by a grating spectrograph and a fast photomultiplier tube (rise-time < 2 ns) by using a beam splitter. The spectrograph, integrating the soot radiation during the first 5 ns after the laser pulse, was attached to an ICCD-camera and the Tire-LII signal was filtered with a BG 1 filter before detection by the PMT, which was attached to an oscilloscope (300 MHz).

Experimental results

No interference with Swan-band LIF emission appeared from the recorded spectrum. The time-decay behavior is influenced by the ambient pressure [1]. In the analysis of the data it appeared that the response time of the system was not short enough to follow the decay rate of the LII radiation. Therefore, a Wiener deconvolution [2] was applied with the response time of the detection system, which also filtered out the noise of the system.

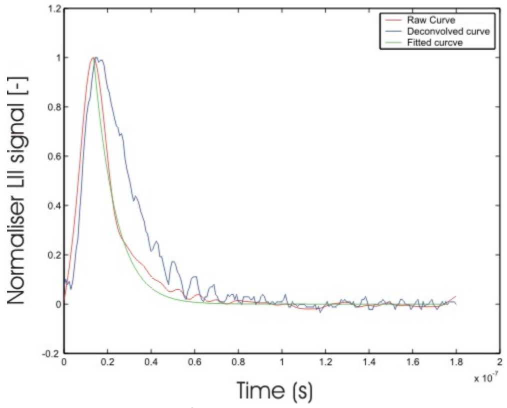

Figure 2: Example of a raw data, a deconvolved curve and a fitted curve according to the model of [3].

A physical model for the cooling behavior of the particles proposed by Kock *et al.* [3] was used to estimate the particle size from the deconvolved data. A fitted curve is shown in Fig. 2 as well. Assuming a mono-disperse particle size distribution, the mean particle size was found to be around 50 nm.

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Modeling of time-resolved laser-induced incandescence (TIRE-LII) transients for particle sizing in high-pressure spray combustion environments

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In particle sizing in particulate laden engine combustion processes it is important to not only provide diagnostic techniques applicable for exhaust gas analysis, but also enable *in-situ* measurements in the combustion chamber under conditions of high pressure and temperature. In a comparative study we present evaluations of signal traces from time-resolved laser-induced incandescence (TIRE-LII) measurements obtained during Diesel spray combustion in a high-pressure combustion vessel as well as from an optically accessible Diesel engine. Results from three currently available LII submodels coupled with a least-squares fitting routine and a “quick-fit” interpolation are compared to retrieve ensemble mean soot particle diameters in a pressure range of 0.6-7 MPa.

Introduction

Recently, the interest has grown in the determination of the amount and size of particulate matter during engine combustion processes due to increased emission regulation standards worldwide. In order to reduce particulate exhaust emissions in Diesel engines a better understanding of in-cylinder soot formation and destruction mechanisms is required. Partly, this task can be accomplished using non-intrusive optical techniques for the in-situ characterisation of soot properties with high temporal and spatial resolution, such as mean particle size and spatial distribution of the soot volume fraction. In laser-induced incandescence (LII) [1] particulate matter is heated to high temperatures via absorption of pulsed laser radiation, and its incandescence radiation is subsequently recorded by zero- or two-dimensional detectors. This allows temporally- and spatially-resolved in-cylinder as well as exhaust-stream diagnostics of soot. In time-resolved LII (TIRE-LII) [2] the soot particle size distribution is reflected in the recorded spectral and temporal characteristics of emitted blackbody radiation during and after laser heat-up. Therefore, TIRE-LII can be applied to retrieve ensemble averaged particle diameters in the measurement volume [3] from fitting temporal signal decays with an appropriate model function derived from the particle energy and mass balance during the heating and cooling phases.

Models

Here, we present a comparison of several data evaluation procedures for TIRE-LII signal transients using three similar physical models currently available in the literature [4-6]. Ensemble mean particle diameters are evaluated through least-squares fitting of model functions to experimentally obtained TIRE-LII signatures. In addition, a “quick-fit” technique, recently proposed by Dankers et al. [7], reduces computational time by fitting single exponentials within two temporal windows of the LII signal trace and extracting mean particle diameters by interpolation in a pre-calculated library of decay times for a range of particle sizes and ambient pressure and temperature conditions.

Experimental results

Experiments were conducted at pressures between 0.6 and 1.4 MPa in a high-temperature high-pressure spray combustion vessel at PSI, and at three crank angle positions in a DI Diesel engine at the University of Nijmegen, where it was shown that even at pressures as high as 7 MPa TIRE-LII signal decays still can be temporally resolved with fast photodetectors. The characteristic parameters of the various models are presented together with preliminary results of mean soot particle diameters using identical data sets obtained from TIRE-LII measurements in high-pressure Diesel spray combustion. The benefits and drawbacks of the data evaluation procedures are discussed.

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Application of TR-LII for the study of carbon vapor condensation at room temperature

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Time-resolved laser-induced incandescence (LII) was applied for the investigation of carbon particle formation at room temperature. A supersaturated atomic carbon vapor was generated by laser photolysis of carbon suboxide. The growth of carbon particles was observed at 0.02 – 1 ms after photolysis. Values of thermal accommodation coefficients of Ar, He, CO and C_3O_2 molecules on the carbon particles surface were determined by the comparison of TR-LII particle sizes with data from Transmission Electron Microscopy.

Introduction

The application of LII to particle-size measurements to various conditions is in the focus of many investigations. Major uncertainties in the interpretation of LII data arise from unknown particle properties like absorption coefficient, density, heat capacity and thermal energy accommodation coefficient of the bath gas molecules on the particle surface. The basic goal of this study is to combine TR-LII technique with sampling measurements to get reliable data about the sizes of carbon particles during carbon vapor condensation at room temperature.

Experimental

Carbon nanoparticle synthesis from laser-induced photo-dissociation of carbon suboxide (C_3O_2) [1] was applied for the study of carbon condensation. A quartz cell (10×10×5 mm^3) was evacuated and filled with pure C_3O_2 or C_3O_2 diluted by Ar, He or CO, respectively, at variable mixture ratios and total pressures in the range from 10 mbar to 1 bar. For photodissociation of C_3O_2 radiation from an ArF excimer laser (193 nm) was used. The energy of each laser pulse was measured in front of and behind the cell to evaluate the energy absorbed by C_3O_2 molecules. The resulting C-atom concentration after photo dissociation was 1.15 – 3.4 $\times 10^{22}$ m^{-3}.

The equipment for the TR-LII measurements consisted of a pulsed Nd:YAG laser (1064 nm) and two fast photo-multipliers for the detection of particle emission at 694 and 550 nm. The laser beam profile was a disc with a diameter of 1 mm and its energy was measured by a calibrated photo-diode for each individual pulse. The energy density of the Nd:YAG laser pulse was less then 0.4 W/m^2 to minimize evaporation of particulate material. LII signals were evaluated using a simple model [2]. To get the particle samples, the TEM grids were deposited on the bottom of the quartz cell and the particles were collected from gas phase under natural gravity conditions for following analysis.

Results

Particle sizes as a function of time after photolysis were determined from TR-LII measurements for different vapor concentrations and gas-diluter pressures (Fig.1). Absolute particle sizes, determined with the assumption of a thermal energy accommodation coefficient α = 1, yielded maximum particle sizes significantly larger than the primary particle sizes observed by TEM.

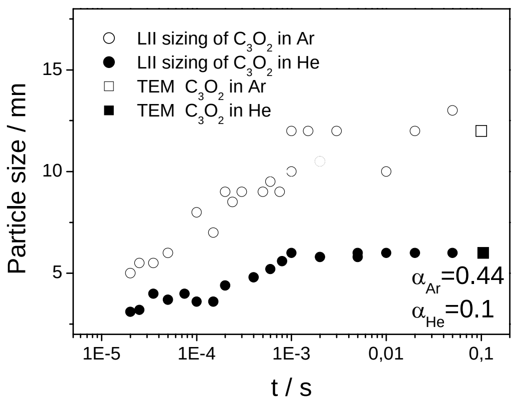

Fig. 1: Carbon-nanoparticle growth in Ar and He. Fit of the TR-LII and TEM particle sizing by thermal energy accommodation coefficients.

The difference between LII and TEM data was attributed to the variations in thermal energy accommodation coefficient α in the different bath gases. Thus, the values of α were obtained for Ar, C_3O_2, He and CO from the comparison of final TR-LII particle size with TEM data.

Gas	Ar	CO	C_3O_2	He
α	0.44	0.44	0.51	0.1

This work is supported by RFBR, DGF and INTAS.

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Planar laser-induced incandescence of iron particles in welding fume

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This paper presents the application of laser-induced incandescence to the measurement of metal and metal-oxide particles resulting from arc welding. Laser-induced incandescence measurements were combined with other optical techniques to investigate spatial distribution and density of particles produced from two arc welding processes. The experimental setup used is outlined along with the effect of different optical variables critical in developing applied optical measurements.

Introduction

Today there is an increased importance being placed on health and safety in the work place. This can be attributed to both the economic value and legislative necessity of maintaining healthy staff. Occupational hazards come in many forms, including electrical, physical, acoustical and chemical. In many occupations such as welding, workers are exposed to a combination of several hazards. One of these is the exposure to fine metal particles contained within the welding fume.

Traditional methods for studying these fumes involve welding within a confined space and collecting these particles in filter paper. While this method is important in determining a fume formation rate it however provides little information in the spatial distribution of the plume. Since the welders are in close proximity to the arc the distribution of the plume is an important factor in determining the welder exposure and inhalation of the fume.

The objective of this work was to apply laser-induced incandescence (LII) to study the spatial distribution and density of metal particles in the atmospheric generated by arc welding process. Two types of welding processes were studied, gas metal arc welding (GMAW) and flux cord arc welding (FCAW). The particle densities were varied by changing the welding conditions, namely the current and the voltage and the flow rate of shielding gas used in the welding process.

The LII signal intensity, as a function of laser fluence, was measured for the fundamental and second harmonic Nd:YAG laser wavelengths. The effect of using different optical colour filters on the LII signal was investigated. 2-d LII images of several gas flows were recorded. Furthermore, 2d images MIE scattering and LII images of the metal particles were collected simultaneously for selected welding conditions. In addition laser extension measurements were preformed aiming to obtain quantitative particles density. Typical LII image of metal particles generated in GMAW welding process is showing in Figure 1 below.

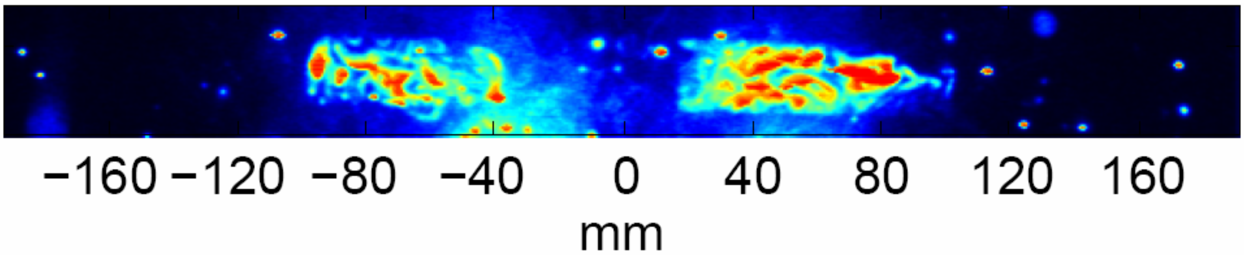

Fig. 1: LII image of GMAW plume cross-section 100 mm above arc.

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Time-resolved laser-induced-incandescence (TR-LII) for sizing of iron nano particles

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Time-resolved laser-induced incandescence (TR-LII) as well as rapid particle sampling and transmission electron microscopy (TEM) were applied to the size measurement of chain-like iron nanoparticles. From the measured TR-LII signals both, the thermal energy accommodation coefficient α_T and the mean particle diameter were determined. The TR-LII-measured particle size is in an excellent agreement with the TEM-determined primary particle size.

Introduction

The objective of the present work is to apply the TR-LII method to the size measurement of iron nano particles. To eliminate the influence of unknown optical properties, the particle temperature after laser heat-up was directly measured by two-color pyrometry. The model used for signal evaluation is described in [1].

Experiments

Iron particles are synthesized in a hot-wall flow reactor, where gaseous iron penta-carbonyl ($Fe(CO)_5$) is used as a precursor diluted in argon and nitrogen, respectively. Within the reactor, the precursor decomposes and nucleation of iron particles takes place [2]. At the exit of the reaction tube, the gas-particle-flow enters an optically accessible measurement chamber. The particles are heated by a pulsed Nd:YAG laser at a wavelength of 1064 nm. The signal is detected perpendicular to the laser beam via a beam splitter and two narrow (10 nm FWHM) band-pass filters at 550 and 694 nm in front of two fast photomultipliers. For particle-size evaluation, the time-resolved signal detected at 550 nm was used, while the signal at 694 nm is used to determine the initial particle temperature by two-color pyrometry. A pneumatic sampling system [3] allows to extract particle probes for TEM analysis. The experiments presented here were performed at p = 500 mbar and T = 670 K with 3200 ppm iron penta-carbonyl. Under these conditions, the magnetic properties of the iron particles cause the formation of chainlike particles [2].

Results

A series of normalized particle emission signals during particle cooling obtained from iron particles in argon with various laser energy densities is shown in fig 1. Although the measurements were performed at identical particle structure and gas conditions, the cooling behavior is obviously different. For an evaluation of the signals in terms of particle size, at first the translational thermal accommodation coefficient α_T had to be determined. For this task, α_T was used as an additional fit parameter together with the mean particle diameter during signal evaluation [1]. In both dilution gases α_T = 0.13 was determined for the iron particles.

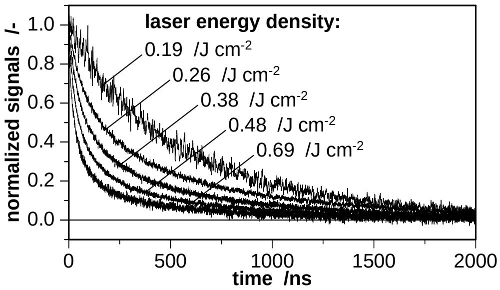

Fig. 1: Normalized TR-LII signals.

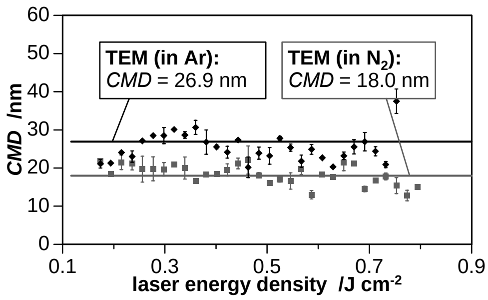

Fig. 2: TR-LII determined particle diameter for iron particles in argon (black) and in nitrogen (gray).

Fig. 2 shows the resulting mean particle diameters as a function of laser-energy density for iron particles in argon or nitrogen, respectively. The continuous lines give the results of the TEM analysis. The TR-LII determined particle size is obviously independent from the laser energy density and is in an excellent agreement with the TEM determined mean primary particle size.

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Laser-induced incandescence of free and surface-adsorbed particles

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The technique of laser-induced incandescence (LII) has been used to study non-soot, in particular silver particles. Experiments were based on experience with LII soot measurements and a sensitivity analysis regarding the influence of various parameters. Some experimental modifications, e.g. double laser pulse excitation, have been tested. A major extension of the LII technique consists in measurements of wall-adsorbed particles requiring appropriate models for data interpretation and evaluation.

Introduction

From its beginning the development of laser-induced incandescence (LII) as an in-situ diagnostics tool was mainly targeted on soot particles. Although some issues of soot measurements are still to be discussed and improved, LII can now be regarded as a standard technique which is widely used in combustion research of both laboratory flames and practical combustion systems (e.g. I.C. engines).

In comparison to soot relatively few studies have been dedicated to other types of particles. This may be surprising as the availability of a well-established technique for measurements on small particles with a good resolution in both, time and location, is of importance in the monitoring of formation or generation and the development and transport of other nano- or micro-sized particles (e.g. TiO_2, ZnO, Ag). Environmental conditions in the production processes of the nanoparticles may change drastically. The physical behavior of the particles on incoming radiation and the heat transfer to the surrounding differs through optical material properties and morphology as well as the surrounding medium. We have investigated the applicability of LII to solid particles and describe results for silver (Ag) particles as an example.

Furthermore we have extended LII experiments to the case where particles are adsorbed on a surface, in our case on a glass plate. Obviously this is more complex than the usual case of a group of freely moving individual particles as the wall attachment leads to additional heat transfer, natural or laser-induced agglomeration, structural deforming etc. Nevertheless there are practical situations where LII measurements on wall-attached particles may be of interest.

Experimental and Modelling Approach

Before beginning with our attempts to extend the LII technique we have checked our experimental set-up and data evaluation by conventional soot measurements. We used standard physical models and parameters which we assessed by a sensitivity analysis. Our experiments on soot particles included their dispersion in different atmospheres (air, argon) and their behavior towards double laser pulse excitation.

In a next step we have set up LII experiments for investigating silver particles. Free particles were excited and observed by using ultrasonic sonotrodes for their local stabilization.

Finally, the LII signals of silver particles adsorbed on a glass surface were investigated. A reasonable data evaluation has to be based on a model extended to describe the considerably more complex situation.

In addition some comparisons with data given by a scanning mobility particle sizer (SMPS) have been made. Furthermore, we investigated the influence of double laser pulses on LII signals as well as the effect of laser illumination on the adsorbed particles.

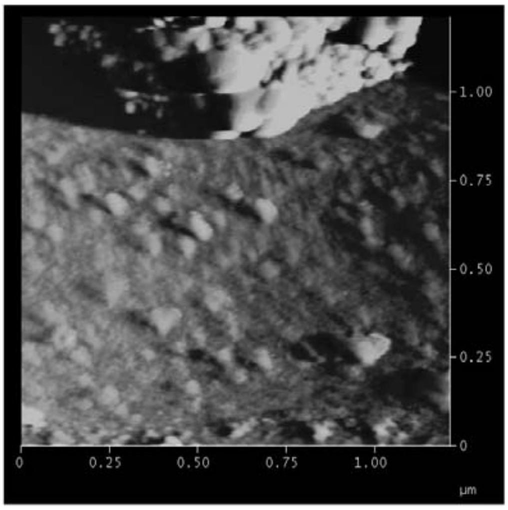

Fig. 1: Atomic force microscopic (AFM) view on the surface of a glass plate with silver particles of 30 nm and their agglomerations of typically 500 nm in diameter

Experimental results and underlying model assumptions are discussed with respect to their accuracy. Some of the drawn conclusions may be regarded to be quite reliable, while others encourage further studies.

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In-situ determination of gas-to-particle reaction generated nanoscaled particles

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One method to determine particle size distributions of nanoscaled particles relies on the simultaneous detection of the time resolved laser-induced incandescence signal (LII) at two different wavelengths. Due to the absorption of a short highly-intensive laser pulse the particles are heated up far above the ambient gas temperature. After the laser pulse the particles are cooling down due to heat transfer, radiation and evaporation. The time-resolved LII-signal describes the temporal evolution of the enhanced thermal radiation. With respect to Planck's law the particle temperature decay can exactly be calculated from the ratio of the time resolved LII-signals at two different wavelengths.

The experimental determination of the particle temperature is based on a developed optical system which separates the induced LII-signal into two wavelength regimes. Two interference filters with a centre wavelength of 650 nm and 450 nm, respectively, filter the separated light beams before they enter a Streak-camera. This is a highly temporally (max. 4 ps) and spatially (~ 35µm) resolving detection system for light intensity measurements. The possibility to detect the time-resolved LII-signal simultaneous with one detection system is one of the advantages of this measuring technique. An improved LII-model with prior validated parameters was used to determine particle-size distributions from measured LII-signals using non-linear multidimensional regression. Knowing the particle temperature for every moment of the cooling phase due to the simultaneous detection of the time resolved LII-signal at two wavelengths it is not necessary to consider the absorption term in the LII model. Thus, there is no need to explain the very well known difference in the maximum particle temperature between the numerical simulated and measured temporal temperature evolution due to uncertainties in the absorption model. In consequence the total uncertainties in the LII-model are less and in comparison to the conventional single-colour method the determined particle size distributions of the two colour method are more correct. The two colour method is a possibility to determine particle size distributions without knowing the optical properties as far as the refractive index is not depending on the wavelength in the regime of 450 nm to 650 nm. Therefore, the knowledge of the refractive index is dispensable and this technique could be adapted to various materials. Determinations of particle size distributions of manganese and iron oxide particles confirmed the applicability of this measuring technique for nanoscaled oxide particles.

One single frequency-doubled Nd:YAG laser pulse was used to determine highly spatially resolved particle-size distributions in the measured volume during gas-phase synthesis of nanoscaled particles. The influence of process parameters on particle formation and growth and therefore on the size distribution could be recognized immediately. A structural characterisation of aerosols with locally different particle size distributions is feasible. Therefore, a possible application for this measuring technique is the usage for air pollution control tasks. Furthermore, it assists to design processes of combustion devices with lower particle emissions or gas-to-particle conversion reactors for the synthesis of nanoscaled particles.

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Two-dimensional imaging of soot volume fraction and OH in turbulent jet diffusion flames spanning low to high mixing rates

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Simultaneous planar measurements of laser-induced incandescence (LII) and Laser-Induced Fluorescence of OH radical were carried out in turbulent jet flames from a simple jet, precessing jet and bluff body burner firing natural gas. These flames span a wide range of global mixing rates and sooting characteristics, and are selected because measurements of total NOx emissions, radiant fraction and global residence time are available. The paper focuses on the choice of experimental conditions required to obtain reliable measurements. However it also discusses key differences in the flames that are obtained.

Introduction

Soot formation and destruction in flames is of great significance due to its role in radiative heat transfer and hence its effect on flame temperature. The development of the Laser Induced Incandescence (LII) technique has enabled experimentalists of obtaining instantaneous two-dimensional images of soot volume fraction in turbulent environments with good temporal and spatial resolution. However the unsteady nature of these flames, combined with the coupled dependence of temperature, radiation, mixing rates and soot volume fraction mean that a detailed understanding also requires simultaneous information of other parameters. Unfortunately such simultaneous measurements are much more difficult in flames containing soot than in clean flames. This is because the scattering from soot particles and the fluorescence from soot precursors contaminate the signal used in the measurement of other parameters.

The aim of the present investigation is to image the location of soot sheets relative to the high-temperature reaction zone on an instantaneous basis. The OH radical, which forms on the fuel-lean side of the high-temperature zone, is deemed to be a reasonable marker for this purpose. Hence, measurement of LII is performed simultaneously with imaging of OH by laser Induced Fluorescence (LIF). The burners selected for the investigation are a simple jet flame, a bluff body flame, which is highly strained, and a precessing jet flame which produces a low strain flame dominated by buoyancy. These span a wide range of mean and instantaneous soot volume fraction at constant total heat output, and have practical relevance [1].

Experiment

LII imaging was conducted using the fundamental from a Nd:YAG laser (1064 nm) to avoid any interference of PAH LIF from soot precursors. The IR laser beam was expanded into a slightly diverging sheet of 85 mm width and a thickness of ~400 µm. This configuration was chosen to maximise the width from a 50 mm lens to facilitate the calculation of soot sheet dimensions, while ensuring that all of the sheet remained within the "plateau region" of the LII fluence curve. The OH radicals were excited with a 282.323 nm sheet from a tuneable Nd:YAG pumped-dye laser producing ~3 mJ/pulse. The two beams were carefully aligned to pass through the same optical train, although the wavelength dependence of the optics means that UV sheet is slightly larger and thinner than the IR. The LII and OH signals were collected on two intensified gated CCD cameras positioned diametrically opposite and perpendicular to the plane of the overlapped laser sheets. To reduce the interference of the OH signal from scattering, a narrow band interference filter with a centre wavelength at 310 nm was used in combination with a long pass glass filter with a cut-off wavelength at about 295 nm, A F1.2 UV lens was used to provide a strong OH signal. Both signals were detected with a gate width of 50 ns, with the LII delayed by 200 ns from the OH.

The paper will report on an assessment of the effects of soot volume fraction and Reynolds number on beam steering for both wavelengths. Since both diffraction and scattering are wavelength dependent, these effects are different in the two sheets, which affects the spatial coincidence of the two sheets, degrading spatial resolution.

The paper will also report briefly on the range and combinations of filters and fuels assessed in seeking to obtain a good OH signal. The simultaneous measurements become more difficult as soot volume fraction is increased due to the combined effects of increased scattering and an apparent reduction in OH concentration. This apparent reduction is presumably due to the combined effects of reduced temperature and modified chemical interactions.

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Peak soot temperature in laser-induced incandescence measurements

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In order to better understand the processes involved in Laser-Induced Incandescence (LII) technique and consequently to develop a theoretical modeling of the physical and chemical processes involved in nanosecond heating of soot particles, many experimental efforts are still required. In this work, results concerning the value of the maximum soot temperature as obtained at the peak of laser-induced incandescence signal are presented. To this purpose, a two-color LII technique has been applied, together with a calibrated lamp used as the reference source. A value close to 4000 K was obtained in flames of different fuels and for many soot conditions (growth, agglomeration and oxidation).

Introduction

The development of non-intrusive optical techniques is of primary importance to understand soot formation mechanisms in combustion systems. Laser-induced incandescence technique (LII) is a promising tool that allows temporally and spatially resolved characterization of soot in terms of soot volume fraction and particle size. Although many works on LII are reported in literature, both on theoretical modeling and on the experimental approach, efforts are still required for an accurate description of this process. Several discrepancies and uncertainties still prevent a straightforward interpretation of the phenomena involved. Soot refractive index, maximum soot temperature, LII signal dependence on laser irradiance are just a few meaningful points of concern. In this work results are presented about the maximum temperature detected at the incandescence peak. This has been performed on different flames and covering all sooty regions in each flame. Basically, the maximum soot temperature is always found to be close to 4000 K, that is the commonly accepted value for soot vaporization.

Experimental and results

A two-color LII experimental set-up was implemented. A portion of the beam of a Nd:YAG laser (1064 nm) is selected by means of a diaphragm and imaged above the burner mouth by a lens (160 mm). In this way a quite uniform and sharp edged beam cross-section is obtained and checked by a webcam. The temporal behavior of LII signal at two wavelengths (450 and 600 nm) is revealed by a monochromator coupled with a photomultiplier and a fast digital oscilloscope. The most suitable procedure to determine the peak LII signal was determined through a statistical analysis of the signal integrated over different gate widths around the peak. The dependence of the peak LII signals at the two wavelengths, and, consequently, of the temperature, on the laser fluence was also investigated. Temperature linearly increases with the laser fluence up to a threshold value (about 250 – 300 mJ/cm^2). With the beam cross-section used, above this value, it remains quite constant with only a slight decrease for laser fluences beyond 600 mJ/cm^2, where vaporization takes place. LII measurements have been performed at about 300 mJ/cm^2, above the threshold level and where vaporization is minimized. Comparison of LII signals with the light intensities from a calibrated tungsten ribbon lamp allowed to determine the LII peak temperature and soot volume fraction by applying well-known two-color emission relationships. Different flames and different soot stages (from inception to oxidation) have been investigated. Fig. 1 shows an example of soot volume fraction and peak temperature axial profiles for diffusion flames fueled with ethylene and methane. As it can be seen, whatever is the soot load and sooty region in each flame, peak temperature is always close to 4000 K. Similar results have been obtained in radial profiles as well.

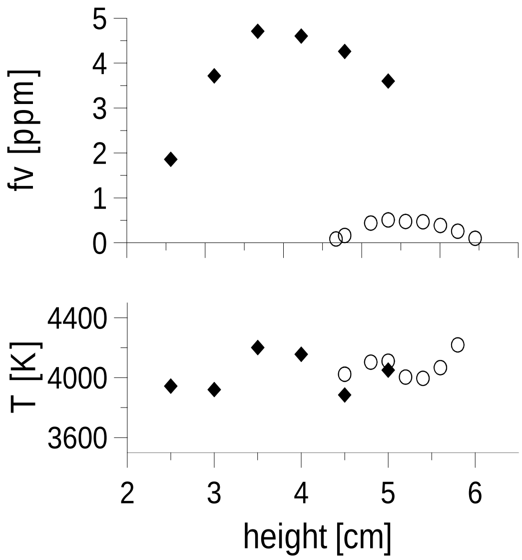

Fig. 1: T and f_v axial profile in C_2H_4 (closed diamonds) and CH_4 (open circles) diffusion flames

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Proceedings of the International Bunsen Discussion Meeting 2005: Laser-induced Incandescence, Quantitative interpretation, modelling, application

Soot volume fractions and primary particle size estimations by means of simultaneous time-resolved and 2D laser-induced incandescence

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The challenge of the present study is to characterize the soot formation "as quantitatively as possible". Due to its good spatial and temporal resolution, the Laser-Induced Incandescence (LII) technique seemed to us to be the most suitable one to quantify the soot. A simultaneous application of two-color time-resolved LII and 2D LII was our particular approach to have in the same time bi-dimensional information regarding soot concentration and punctual information on particle mean size. A pilot study was conducted in order to quantify the soot volume fractions and particle size in an atmospheric pressure laminar flame by means of this simultaneous technique. Soot concentration and size estimations have been eventually performed inside the combustion chamber of a Diesel engine.

Introduction

The combustion characteristic to Diesel engines is a source of pollutant emissions, the soot being one of the most important representatives. Even that the latest technologies e.g. high-pressure direct injection have a direct contribution to the Diesel soot emissions reduction, further research should be done to meet the limits imposed by the pollution limits in order to preserve the air quality. Before finding solutions to reduce the pollutant emissions, it is necessary to understand their formation and, if possible, to perform quantitative *in-situ* measurements.

Laser-Induced Incandescence Application

One of the techniques that allow quantifying the soot non-intrusively and in situ is Laser-Induced Incandescence (LII), this technique being in our opinion the most suitable to apply for soot volume fraction and the primary particle mean diameters estimations inside the combustion chamber. It consists basically in heating the particles by means of a pulsed laser beam up to the vaporization temperature and to record the incandescence signal that has a diameter-dependent evolution versus time and scales with the soot amount in the measurement volume.

One of this technique's most recent approaches is based on estimating the soot heated temperature by means of the two-color pyrometry. It is based on the recording LII signal at two wavelengths. The soot volume fraction is then calculated with a Planck-type formula [1]. The individual time-resolved (TR) signals may be used to estimate soot particle sizes trough energety transfer models.

A combination of the two-color TR-LII technique and 2D LII imaging allows us to make estimations regarding the soot concentration inside the combustion chamber of an optically accessible Diesel engine. Our strategy consists in recording a time-resolved signal filtered at two different wavelengths simultaneously with a LII image. The two-color time-resolved signals recorded for a small measurement volume allow estimating the soot laser-heated temperature and then the soot volume fraction for that "punctual" location. The ratio between the so-calculated soot volume fraction and the average image intensity corresponding to the small measurement volume is actually the factor that we use to express the intensity units of the image in soot volume fraction units.

The experimental set-up consists of a pulsed Nd:YAG laser at 1064 nm, the reactive media (optically accessible Diesel engine or atmospheric pressure flame) and for signal detection and recording an ICCD camera and two photomultipliers tubes connected at a rapid acquisition system.

Soot volume fractions and soot sizes in the atmospheric pressure flame were first estimated with the two-color LII technique in several points of the flame. Our simultaneous application of the LII technique in the flame proved a good agreement with the two-color pyrometry results. Soot size estimation in atmospheric conditions has been done through the LII model described in the reference [2]. Thermophoretic sampling results prove that the estimated mean diameter assuming monodisperse soot size is close to the count mean diameter of the lognormally-distributed sampled soot size. Soot volume fractions and sizes were eventually estimated inside the combustion chamber of a Diesel engine. The soot size was estimated through the high pressure model described in reference [3]. EGR effects on soot formation were also investigated.

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Time-resolved laser-induced incandescence applied to in-cylinder Diesel particle sizing

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Laser-induced incandescence with time-resolved detection (TR-LII) in two frequency channels was established and applied to the combustion chamber of a single-cylinder two-stroke Diesel engine. It was possible to determine the mean particle diameter and the geometric standard deviation of an assumed lognormal size distribution for the in-cylinder particles as a function of the crank angle. The measured sizes were in good agreement with the primary particle sizes in the exhaust gas determined by TEM-analysis.

Introduction

For any further reduction of the particlulate emissions of Diesel engines a better understanding of the processes of particle formation and oxidation inside the combustion chamber would be helpful. For that purpose a particle-measuring method yielding instantaneous information about the particle size distribution at different stages of the combustion cycle would be worthwhile.

Time-Resolved Laser-induced incandescence (TR-LII) has become a common method for in-situ particle size measurements. In the present study a two-color version of TR-LII [1, 2] was established and applied to a Diesel engine. The objective was to determine the particle count median diameter *CMD* and geometric standard deviation σ_g of an assumed lognormal size distribution of the particles as a function of the crank angle.

Experiments

The experiments were performed on a single-cylinder, two-stroke Diesel engine with a displacement volume of 250 cm^3. A custom designed cylinder head provides the required optical access. The engine was motored by an electrical asynchronous motor at a constant speed of 1500 min^{-1}. For the TR-LII measurements, the engine was fired for some individual cycles only. All experiments were performed at an injection crank angle of 23° CA BTDC and an air/fuel equivalence ratio of $\phi = 0.26$. The particles were heated by an Nd:YAG laser at 1064 nm with a laser fluence of 0.10 J/cm^2.

Finally, a thermophoretic particle sampler was located in the exhaust gas manifold to get particle probes for further analysis by transmission electron microscopy (TEM).

Results

A series of normalized particle emission signals during cooling obtained at six different crank angles is shown in Fig. 1. Particle cooling strongly depends on the engine crank angle and becomes longer with increasing crank angle.

The signals of Fig. 1 and others were evaluated in terms of particle size by fitting lognormal distribution functions to the measured curves under the variation of *CMD* and σ_g. The results are shown in

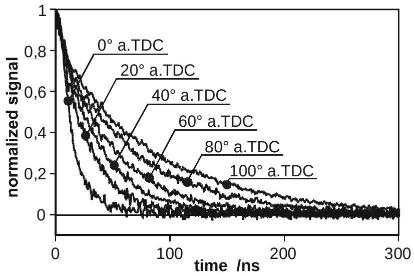

Fig. 1: Normalized TR-LII signals at six crank angles.

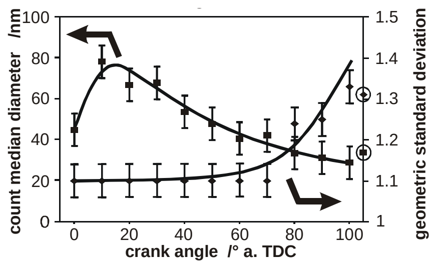

Fig. 2: CMD and σ_g evaluated from the signals in Fig. 1.

Fig. 2. *CMD* is in the range of 30 to 75 nm, increases up to a crank angle of about 10° CA and decreases again towards a value of about 30 nm at 100°CA after TDC. This behavior can be explained by particle formation and subsequent particle oxidation. σ_g is constant at a value of 1.1 up to a crank angle of 70°CA and then increases towards about 1.32. The two circled values of *CMD* and σ_g, which are shown at the right edge of the diagram, are results of the TEM analysis of the exhaust gas primary particles. The agreement between the TR-LII measured sizes at 100°CA after TDC and the TEM determined primary particle sizes in the exhaust gas is quite good.

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Gas-phase temperature imaging in sooting flames by multi-line NO-LIF thermometry

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Two-dimensional gas-phase temperature fields were measured in sooting atmospheric pressure and elevated pressure flames with multi-line NO-LIF thermometry. The data is then used as input for modeling particle sizes with temporally resolved LII measurements at high pressure. We also compare flame temperature values derived by pyrometry measurements with the gas-phase temperatures measured in a stabilized McKenna flat flame and find some differences in the lower part of the flame.

Introduction

The gas-phase temperature in flames is of high interest because it governs chemical reaction kinetics and influences the LII signal decay rate significantly [1].

Multi-line NO-LIF thermometry

In contrast to conventional two-color LIF thermometry for gas-temperature the multi-line technique yields absolute temperatures without calibration [2] and the technique can be applied even in systems with strong scattering and fluorescence background. The technique is based on the measurement of LIF (Laser Induced Fluorescence) excitation spectra of nitric oxide (NO). Some thousand ppm of NO are added to the fresh gas as a fluorescent tracer. The laser is tuned over a part of the NO absorption spectra while individual images are taken with an intensified CCD camera for each excitation wavelength. From the resulting stack of pictures (each with the laser tuned to the next wavelength) LIF excitation spectra can be extracted for each pixel. Simulated spectra are then fit to the experimental data using LIFSim (www.lifsim.com). Free parameters are absolute temperature, broad-band background as baseline and signal intensity.

A tunable KrF*-excimer laser (248 nm) is frequency-shifted to 225 nm in an 8-bar H_2 Raman cell. The laser beam is formed to a 40 × 0.5 mm^2 light sheet which illuminates a vertical plane in the region of interest. The NO molecules are excited in the A-X(0,0) band and the LIF-signal is recorded with an intensified CCD-camera. Elastically scattered light is suppressed by Schott UG5 filters. An additional reflection band pass filter separates the 230 – 255 nm range for detection to suppress fluorescence from soot precursor molecules.

High pressure flat flame burner

The gas phase temperature could be determined in an elevated pressure ethylene flat flame with an equivalence ratio ϕ = 2.1. For 1 bar we find 1644 ±30 K, for 2 bar 1710 ±20 K and at 5 bar we measured 1850 ±100 K in the middle of the flame 10 mm above the burner matrix. An excitation spectrum and the fit is shown in Fig. 1. The data is used as input for modeling particle sizes with LII.

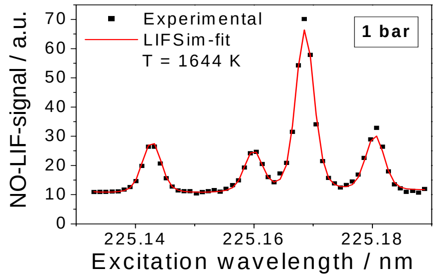

Fig. 1: NO-LIF excitation spectrum at 1 bar in a sooting ethylene/air flame at ϕ = 2.1

Soot particle versus gas-phase temperature

The multi-line NO-LIF technique has been used to obtain temperature fields (Fig. 2) of sooting atmospheric pressure flat flames on a McKenna burner stabilized by a metal plate. Soot particle temperatures derived by pyrometry [3] showed deviation from the gas-phase temperature in the lower flame zone.

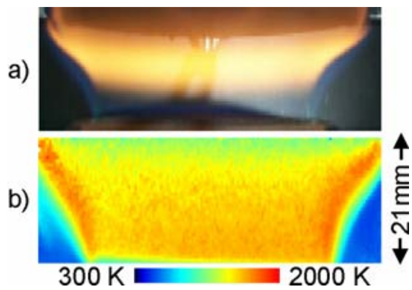

Fig. 2: a) Photograph and b) gas-phase temperature field of a sooting flame stabilized on a McKenna burner

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A critical evaluation of the thermal accommodation coefficient of soot determined by the laser-induced incandescence technique

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Data of several laser-induced incandescence (LII) experiments reported in the literature were re-analyzed to obtain the values of the thermal accommodation coefficient (TAC) of soot. The present analysis shows that the TAC of soot falls in a relatively narrow range between 0.23 (in N_2 and flames) and 0.45 (in Argon). Using an aggregate based low-fluence LII model, the TAC of soot in a laminar diffusion flame was found to be 0.38.

Introduction

Knowledge of TAC of carbonaceous materials is critical in the determination of primary particle diameter of soot and carbon black. Conversely, LII can be used to determine the value of TAC of soot [1-4]. Unfortunately, the reported values of TAC of soot vary over a wide range from 0.07 [1] to 1 [2].

The objective of this study is to re-analyze the experimental data reported in [1-4] for the purpose of extracting the TAC of soot.

LII Model

The LII model used in this study was that developed at NRC. The model simulates the entire temperature history of soot particles. Unless otherwise stated, the single primary particle version of the model was used in the evaluation. The heat conduction sub-model used is the Fuch's approach.

Results

The particle temperature data of the lowest fluence case shown in Fig.3 of [1] was used to extract of TAC of soot in the atmospheric pressure premixed flame. Fig. 1 shows the temperature history and the best estimate of TAC of soot is 0.23 rather than 0.07 [1].

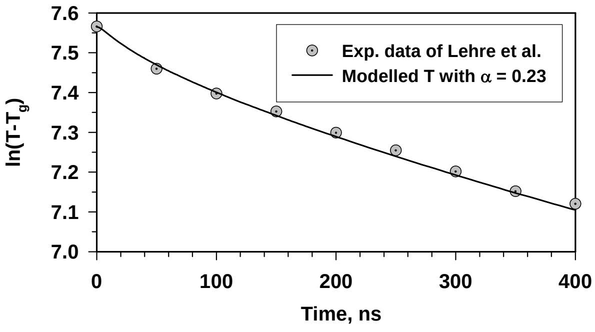

Fig. 1 Comparison of particle temperature.

The normalized LII signal shown in Fig.3 of [2] was calculated using the present LII model and the results are shown in Fig. 2. The TAC of soot in Argon was found to be 0.45, not 1 [2].

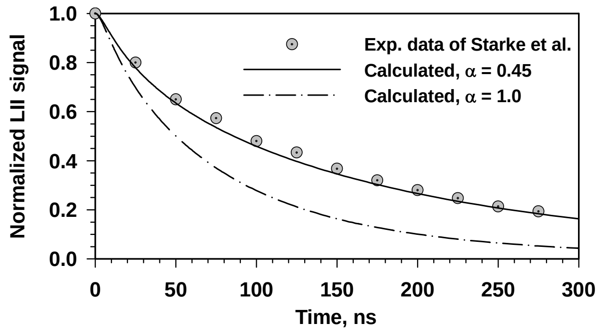

Fig. 2 Comparison of normalized LII signal.

The study of Kuhlmann et al. [4] employed high-fluence laser. The found the TAC of soot to be 0.23. High-fluence LII suffers the problem of soot sublimation which reduces the soot particle diameter. As a result, the TAC of soot is subject to large uncertainty unless the particle diameter at the end of soot sublimation is available, see Fig.3.

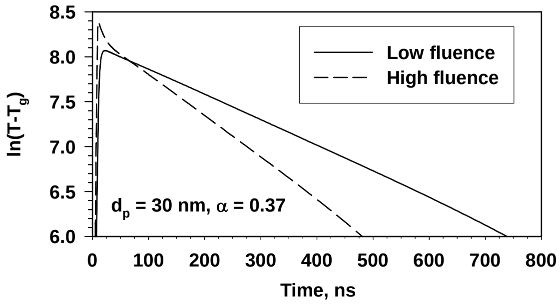

Fig. 3 Particle temperature decay: T_g = 300 K, p = 1 atm.

Application of an aggregate based low-fluence LII to the data in [3] obtained the TAC of soot of 0.38.

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