Fuelling a cleaner future : Quantification and characterization of soot emissions from combustion based energy conversion - Year 3 Report

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**PROJECT ASSESSMENT FORM/
FICHE D'EVALUATION DES PROJETS**

DATE: 2011.04.18

PROJECT TITLE/TITRE DU PROJET

Fuelling a cleaner future: Quantification and characterization of soot emissions from combustion based energy conversion

PARTNERS/PARTENAIRES

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BRIEF DESCRIPTION OF THE PROJECT AND ITS SCIENTIFIC OBJECTIVES/ BREVE DESCRIPTION DU PROJECT ET DE SES OBJECTIFS SCIENTIFIQUES

Soot emission from industrial combustors and engines has a major detrimental impact on air quality and human health, and is now being recognized as a key factor in global warming. Soot formation also plays an important role in the performance of these technologies. Laser-based measuring techniques are important tools for understanding soot formation and oxidation in flames and for the quantification and characterization of particulate emissions from energy generation, industrial, and mobile sources. Laser-induced incandescence (LII) is an emerging technique that offers many advantages and unique capabilities over existing measurement technologies. However, for this approach to mature into a reliable tool for the determination of particulate concentration and morphology a physical model that accurately describes the nanoscale heat and mass transfer processes in LII must be developed to provide reliable interpretation of the LII experimental data. Integrating LII with other diagnostics will also improve the reliability and broaden the utility of LII for the characterization of complex, transient problems such as measurements for gas turbines.

The three main objectives of the collaboration are:

- address uncertainties in the LII physical model, including the optical properties of soot, the thermal accommodation coefficient, and the compositions of vaporized carbon species during the LII process.
- to optimize and integrate a suite of diagnostics including 2D-LII (two dimensional LII), PIV (particle image velocimetry), and CARS (coherent anti-Stokes Raman spectroscopy) which can be used for the characterization of complex transient sooting flames.
- to establish an experimental database (soot properties, temperature, and velocity) for the evaluation of numerical simulation of soot formation and oxidation, which currently does not exist, from a selection of well-defined sooting flames.

This unique experimental database can be used for the validation of kinetic soot models and numerical flame simulations which could be applied to the development of ultraclean combustion systems.

Specific objectives for the third year of the collaboration included:

- enhance capabilities of optical soot diagnostic techniques based on gaps identified; extend range of flame conditions for measurements performed in first 2 years; improve numerical simulation models using data acquired in first 2 years
- prepare final report

PROJECT STATUS AND DURATION/ETAT DU PROJET ET DUREE EFFECTIVE

Start Date/Date de démarrage	Planned completion date/Date prévue d'achèvement
April 1, 2008	July 31, 2011

FUNDING/FINANCEMENT

	NRC-ICPET		Helmholtz-DLR	
Total funds allocated for project/Total des fonds alloués au projet	$485k		k€ 410	
Funds received/Fonds reçus	$485k		k€ 410	
Funds allocated/Fonds alloués	$485k		k€ 410	
Forecast/Prévision	$0k		k€ 0	
Breakdown of funds expended to date/Ventilation des dépenses engagées à ce jour	Cash	In-kind	Cash	In-kind
Human resources/Resources humaines	$275k	$305k	k€ 300	k€ 240
Equipment/Equipement	$90k	$115k	k€ 15	k€ 30
Operations/Frais de fonctionnement	$54k	$0k	k€ 30	k€ 0
Travel/Frais de déplacement	$51k	$0k	k€ 35	k€ 0
Other/Autres frais	$15k	$0k	k€ 30	k€ 0
Total expenditure/Total de dépenses	$485k	$420k	k€ 410	k€ 270

SCIENTIFIC ACHIEVEMENT/REUSSITES SCIENTIFIQUES

Results already obtained/Résultats d'ores et déjà obtenus:

Year Three

- Characterization of a turbulent non-premixed turbulent flame using a suite of optical diagnostics completed including combined PIV+LII developed during the second year of the collaboration. Published in Applied Physics B. Data used for validation of an in-house combustion model at DLR. Publication in preparation on combining full set of experimental data and modelling for Combustion and Flame.
- Completion of high spatial resolution LII diagnostic. Applied to Gulder burner and data used for improvement of LII model.
- Refinement of heated LOSA; diagnostic used to study the variation of soot optical properties during rapid laser heating. Published in Applied Physics B. Technique extended to allow measurements of in-flame soot. Paper prepared and oral presentation at upcoming Combustion Institute/Canadian Section Spring Technical Meeting
- Study of spectral variation of soot optical properties with wavelength and soot age completed and published in Applied Physics B.

- Apply Generalized Mie Method (GMM) model to investigate effect of nanoparticle aggregation on optical absorption. This demonstrated that the Rayleigh-Debye-Gans (RDG) approach that is commonly used introduces uncertainties. The new approach represents a much more detailed understanding of the light-matter interaction on the nanoscale. Published in the Journal of Quantitative Spectroscopy and Radiative Transfer.

Year Two

- The team has demonstrated a novel combination of LII and TOF-MS (time-of-flight mass spectrometry) to determine the vaporized species during LII heating, drawing on the extensive experience of Dr. Grotheer (DLR) for TOF-MS and Dr. Thomson (NRC) for laser desorption, laser ionization mass spectrometry (LD-LI-MS). The research to date has shown that at intermediate fluences significant volatilization is possible in young soot. Details on the breakdown of soot particles into a new class of small particles and the formation of carbon clusters is critical information for the improvement of soot vaporization models as implemented in soot modelling. Published in Applied Physics B.
- Improvement of existing LII modelling at DLR due to continuous discussions with NRC colleagues.
- Optimization of 2D LII calibration procedure now using 1064 nm as attenuation wavelength, in combination with an improved thermal stability of the detection, integration of the stabilized detectors in the existing mobile attenuation device and of the data acquisition into LabView control. Components and design have been provided to NRC.
- Heated LOSA (line-of-sight attenuation) experiment for monitoring optical soot properties during LII excitation was optimized by Dr. Geigle (DLR) during his research exchange to NRC. Measurements made in the exhaust of a Gülder flame show that soot density must be accounted for during LII signal interpretation. In addition, variation of the optical properties of soot with heating was found to be different for near ultra-violet (UV) and near infrared (NIR) wavelengths.
- Extensive work was performed to extend available data base for standard burners
- CARS temperature data for the McKenna flame obtained and data analysis is ongoing. These experiments were a first test of the new DLR dye laser system integrated into the mobile SV (shifted vibrational)-CARS laser container for performance optimization.
- Spectral LOSA E(m) absorption function variation data in McKenna flame
- TEM (transmission electron microscopy) imaging soot morphology data in McKenna and Gülder flames
- 2D TiRe (time-resolved) LII particle sizes for Gülder burner at DLR, using CARS temperature measurements performed by NRC. In contrast to frequently employed photomultiplier point detection this approach allows for 2D particle size mapping in stationary flames at the intensified camera spatial resolution.
- Simultaneous PIV/LII demonstrated in heavily sooting flames during B. Crosland's research exchange to DLR, data complement data base mentioned below for sooting jet flame
- Two-dimensional, two-colour LII demonstrated in Gülder flame, now published in Applied Physics B
- Turbulent sooting jet flame defined in cooperation with numerics colleagues, data

base of various optically determined flame parameters started. This flame combines the advantages of limited size, significant soot formation, relevant turbulence and simple, well defined geometry suitable for CFD (computational fluid dynamics) modelling.

- 532 nm and 1064 nm LII systems modified to allow high spatial resolution measurements, optimized for detailed measurements in standard flames.
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Year One

- Discovered an elliptical heat transfer relationship for soot conduction cooling, which has not previously been discussed or published. This provides an advancement in the understanding of the energy balance for nanoparticles undergoing temperature fluctuations with rates as high as 1000K/ns.
- Development and demonstration of 2D-AC-LII. The extension of the point measurement approach previously employed in laser-induced incandescence to a two-dimensional imaging approach will require extensive optimization. This first demonstration illustrated that the 2D approach is effective and also highlighted opportunities for improvement.

Remaining objectives/Objectifs restant à atteindre:

- Compilation of all heated LOSA results to understand the impact of heating of soot on soot optical properties and the impact on LII measurements. Peer reviewed publication

OUTCOMES/RESULTATS

Exchanges (originating organization, Senior Researcher, Ph.D student/intern, planned dates and duration)/Echanges (sens de l'echange, chercheur confirmé, thésard/stagiaire, dates et durée prévue)

- K.P. Geigle (DLR, Senior Researcher) at NRC: April/May 2011 (1 month) involved in heated LOSA data interpretation
- M. Köhler (DLR, Post Doctorate) at NRC: summer 2010 (2 months) performed heated LOSA experiments.
- F. Migliorini (NRC, RA) at DLR (Jan-Mar 2010, 2.5 months) continued MS/LII experiments using improved definition of boundary conditions.
- K. Thomson (NRC, AcRO) at DLR (Dec 2009, 1 month) performed joint MS/LII experiments to determine effects of pulsed LII laser excitation on soot particles.
- B. Crosland (NRC, Ph.D. Student) at DLR (Nov-Dec 2009, 2 months) performed simultaneous PIV/LII.
- K.P. Geigle (DLR, Senior Researcher) at NRC (Jul-Oct 2009, 3 months) performed heated LOSA

Joint publications (specify date)/Publications conjointes (préciser les dates)

1. Köhler, M. , Geigle, K.P., Meier, W., Crosland, B.M. , Thomson, K.A., and Smallwood, G.J., "Sooting Turbulent Jet Flame: Characterization and Quantitative Soot Measurements," accepted to Applied Physics B, January 2011, DOI:10.1007/s00340-011-4373-y
2. Thomson, K. A., Geigle, K.P., Köhler, M., Smallwood, G. J., Snelling, D. R., "Optical Properties of Pulse Laser Heated Soot," accepted to Applied Physics B, February

2011, DOI 10.1007/s00340-011-4449-8

3. K.P. Geigle, K.A. Thomson, G.J. Smallwood, G. Zizak, "Synopsis of recent LII research activities as presented on the 4th International Workshop on LII," International Energy Agency 32nd Task Leaders Meeting on Energy Conservation and Emissions Reduction in Combustion, Nara, Japan, 25-29 July 2010.
4. K.P. Geigle, M. Köhler, K.A. Thomson, G.J. Smallwood, D. R. Snelling, "Influence of Laser Heating on Soot Optical Properties," Work in Progress Poster, Combustion Institute, International Symposium, Beijing, China, August 2010
5. Geigle, K.P., Köhler, M., Thomson, K. A., Smallwood, G. J., Snelling, D. R., "Optical Properties of Laser Heated Soot Aggregates," oral presentation and proceedings, International Energy Agency XXXII Task Leaders Meeting on Energy Conservation and Emissions Reduction in Combustion, Nara, Japan, July 2010.
6. Köhler, M., Geigle, K-.P., Meier, W., Crosland, B., Thomson, K. A., Smallwood, G. J., "Sooting turbulent jet flame: characterization and quantitative soot measurements," oral presentation at the 4th International Workshop and Meeting on Laser Induced Incandescence: Quantitative Interpretation, Modeling, Application, April 18-20 2010, Varenna, IT
7. Geigle, K-. P., Thomson, K. A., Smallwood, G. J., Snelling, D. R., "Influence of pulsed laser heating on the optical properties of soot," oral presentation at the 4th International Workshop and Meeting on Laser Induced Incandescence: Quantitative Interpretation, Modeling, Application, April 18-20 2010, Varenna, IT
8. K. Wolf, K.Thomson, F.Migliorini, H.-H. Grotheer, G. Smallwood, M. Köhler, K.P. Geigle, "IR-Laser Ablation of Species from Growing Soot Particles and their Detection through Mass Spectrometry." oral presentation at the 4th International Workshop and Meeting on Laser Induced Incandescence: Quantitative Interpretation, Modeling, Application, April 18-20 2010, Varenna, IT
9. F. Migliorini, K.A. Thomson, K.P. Geigle, G.J. Smallwood, "Investigation of Soot Optical Properties by Spectral Line-of-Sight attenuation", International Energy Agency XXXI Task Leaders Meeting on Energy Conservation and Emissions Reduction in Combustion, Lake Louise, AB, Canada, 20-24 September 2009.
10. F. Migliorini, K.A. Thomson, K.P. Geigle, G.J. Smallwood, "Investigation of Soot Optical Properties by Spectral Line-of-Sight attenuation", Poster presentation, Gordon Research Conference, Laser Diagnostics in Combustion, Waterville Valley, NH, USA, 16-21 August 2009.

Further publications tied to the project but not involving joint authorship have been excluded from this list.

Visits, including joint participation in conferences, workshops, presentations, etc. (specify date)/Visites, incluant la participation commune à des colloques, ateliers et présentations (préciser dates)

- DLR -> NRC July 2011, 2 days, final project wrap-up meeting
- SAE E-31 Aircraft Exhaust Emissions Measurement Committee Annual Meeting, 29 June – 2 July 2010, Interlaken, Switzerland
- 4th International Workshop on Laser-Induced Incandescence, 18-20 April 2010, Varenna, Italy
- NRC -> DLR, December 2009, Greg Smallwood, 2 days, experiment review, discussions, and planning
- International Energy Agency XXXI Task Leaders Meeting on Energy Conservation and Emissions Reduction in Combustion, Lake Louise, AB, Canada, 20-24

September 2009 ▪ DLR -> NRC March 2009, Klaus-Peter Geigle, 2 days, first year meeting ▪ NRC – organizer of 3rd International LII workshop, Ottawa, July 2008. Assisted with program, session chairing, and proceedings by Klaus-Peter Geigle ▪ DLR -> NRC May 2008, Klaus-Peter Geigle & Jochen Zerbs, 3 days, facility visit, experiment planning ▪ NRC -> DLR, March 2008, Greg Smallwood, Fengshan Liu, & Kevin Thomson, 3 days, facility visit, experiment planning
Teaching/Enseignment
Technology transfers, patents and licences, contracts with industry/Transferts de technologie, brevets et licences, contrats avec l'industrie

FUTURE RESEARCH PLANS/PLANS DE RECHERCHE FUTURS

Due to the highly successful outcomes of the collaboration, both sides of the collaboration are highly motivated to find new opportunities for collaborative activities. Financial support is a key ingredient to a successful collaboration and so we will focus on finding a project of industrial relevance where the combined resources of NRC/ICPET and DLR are seen as advantageous – likely involving the aviation sector.

Including the personnel exchange proved extremely valuable for the exchange of experiences, stimulated discussion, and focussed research activities afforded by being a visiting researcher. Our research partners are highly interested in locating financial support for future exchange and intend to maintain the newly created link as much as possible after the end of the current cooperation.

BENEFITS OF COLLABORATION/AVANTAGES DE LA COLLABORATION

The joint research is advancing the state of the science of the nanoscale heat and mass transfer processes in LII, nanoparticle formation and characterization. The exchange and analysis of data obtained from well-defined experiments including electron microscopy, light scattering, multi-wavelength line-of-sight attenuation and emission, and coherent anti-Stokes Raman spectroscopy (CARS) as well as numerical modeling carried out by both organizations is leading to an improved understanding of the nanoparticle formation and oxidation processes in both organizations, and at a rate that would not be possible for either organization to achieve on its own. The outcome is advantageous for both institutions and, in addition, to the scientific community dealing with LII as measurement tool for soot in combustion.

The collaboration has been highly productive for both parties and allowed more rapid progress than could have been anticipated at the onset of the collaboration. We attribute the success to a few key features of the exchange program as well as to the intrinsic benefits of a collaboration of two complementary organizations:

1.) Funding for travel between the organization was targeted in the project plan
    a. good communication and regular face-to-face meetings were integral to maintain a focus on the project deliverables and progress
2.) Research exchanges were emphasized in the program structure, both as a deliverable and in terms of funding to all the exchanges
    a. working with another research team for a short time period allows the rapid acquisition of new research skills and the opportunity to observe how other research teams function and plan. Best practices were identified and brought back to the visitors home research team.
    b. short research exchanges inspired intense research campaigns where a great deal was accomplished in short time periods
    c. sending researchers on exchanges allows them some distance from the bureaucracy which can tend to overwhelm their day-to-day routines in their own organization and allows them to remember and revitalize their research skills
3.) No single laboratory has all the diagnostic tools and expertise to tackle the research tasks which were targeted in this collaboration
    a. complementary diagnostics and a sharing of tasks allowed for greater progress than either organization could achieve acting independently

An additional benefit is that the collaboration with DLR was seen as enhancing ICPET's reputation in the international aviation sector, and attracted the attention of Transport Canada. TC is now funding ICPET research with the goal of developing a new international certification standard for the measurement of nonvolatile particulate matter emissions from aviation engines.

PROBLEMS AND DIFFICULTIES ENCOUNTERED IN THE IMPLEMENTATION OF THE PROJECT/PROBLEMES ET DIFFICULTES RENCONTREES POUR LA MISE EN ŒUVRE DU PROJET

We encountered delays in identifying and hiring a research associate appropriate for the project. It would be helpful if a period of about 6 months between the announcement of successful proposals and the start of the research collaboration period could be built into the program schedule to allow for the hiring of research staff.

OTHER COMMENTS/COMMENTAIRES ADDITIONNELS

ASSESSMENT REVIEWED AND APPROVED BY THE DIRECTOR GENERAL OF (name of NRC institute)/EVALUATION REVUE ET APPROUVEE PAR LE DIRECTEUR GENERAL DE (nom de l'institut)

	2.05.2014
Director General/Directeur général	**Date**