Activity Report from the Division of Combustion Physics 1997-1998

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Cover illustration: PLIF images of OH in a turbulent diffusion flame, acquired in rapid sequence.
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1. INTRODUCTION

The Division of Combustion Physics is since 1991 a separate division within the Department of Physics at Lund Institute of Technology, LTH. The scientific activities have a long tradition in developing and applying laser techniques for combustion diagnostics. There is also a long tradition to work in the field of ignition and spark phenomena, mainly directed towards SI engines. During the last years there has also been strong activities started in the field of theoretical chemical kinetics. Within the combustion modelling activities there is also work in the field of turbulent combustion.

All activities as mentioned above are major parts in the Combustion Centre, which is an interdisciplinary Centre within LTH, with the aim to create links between different disciplines within the Institute in the area of combustion. Thanks to this Centre, the Division has during the last years been heavily involved in national concentrations, e.g. the Centre of Competence in Combustion Processes financed by NUTEK, industry and the University, as well as the National Graduate School in Combustion Science and Technology, CECOST. The laser activities within the Division has also benefitted from being part of the Lund Laser Centre (LLC). LLC constitutes a EU Large Scale Facility and thus the Division has had several European visitors participating in various experiments.

Thanks to Centre activities above, the number of people within the Division has increased by almost 50% in three years. Due to this enlargement of the activities, not only within the Division but also at neighbouring departments working in the field within the Institute, a decision has been taken by the Dean of the Institute to investigate for the constructing of a new building within the Department of Physics. This building, of about 2000 m², is planned to be ready in early 2001 and would host not only all personnel from the Division of Combustion Physics but also persons from the Dept. of Heat and Power Engineering, as well as Dept of Fire and Safety Engineering. Besides the increase of number of persons, an additional reason for the building is a recent grant on 20 MSEK from DESS (Delegationen för Sydsveriges Energiförsörjning) for the construction of a burner with the possibility to study turbulent combustion phenomena at elevated pressure (7-8 bar) and preheated air (~900 K). Within the building there are also plans for additional new burners as well as special laboratories.

The Division has not yet been strongly involved in education at an undergraduate level (MSc). In order to increase the student exposure of the combustion research being made as well as showing the complexities involved in combustion, the Division has been heavily involved in the process of trying to create a special program within the MSc education directed towards combustion science phenomena. Close contacts have also been established with other national technical universities and major Swedish industries in the field to create a national program and collaboration between these universities to facilitate for MSc students to read and study courses within the combustion field at other the universities.

Marcus Aldén, professor
Head of Combustion Physics
2. GENERAL INFORMATION

2.1 Staff

The staff included the following members:

Marcus Aldén, professor, Head of Division
Per-Erik Bengtsson, PhD, Docent
Clemens Kaminski, PhD
Philip Klaus, PhD (971201-)
Fabian Mauss, PhD
Kaj Nyholm, PhD (-980831, present position: Tech University, Helsinki, Finland)
Christer Löfström, res. eng.
Hans Neij, res eng (970401-981130, present position: AWApantent, Malmö)
Per Amnéus, grad. stud. (970701-)
Öivind Andersson, grad. stud.
Boman Axelsson, grad. stud. (971101-)
Michael Balthasar, grad. stud.
Joakim Bood, grad.stud.
Christian Brackmann, grad. stud. (980301-)
Robert Collin, grad.stud. (981001-)
Anne Dederichs, grad.stud (970915-)
Johan Engström, grad.stud.
Axel Franke, grad.stud. (970818-)
Rolf Fritzon, grad.stud. (present position: Kransell & Wennborg AB, Stockholm)
Johan Hult, grad.stud (980301-)
Terese Lövås, grad.stud. (980901-)
Thomas Metz, grad.stud.
Sven-Inge Möller, grad.stud
Daniel Nilsson, grad.stud
Frederik Ossler, grad.stud.
Raymond Reinmann, grad.stud. (-980131, present position: SAAB Automobile AB, Södertälje)
Mattias Richter, grad.stud.
Joachim Walewski, grad.stud (970801-)
André Saitzkoff, amanuens (-980831)
Elna Brodin, secretary
Jörgen Hasth, administrative director CECOST
Birgitta Olofsson, secretary CECOST
Rutger Lorensso, instrument maker
2.2 Visitors

Ulf Bergmann, Tobias Hartlieb, Heidi Böhm, Axel Lamprecht, Guido Horst
Universität Bielefeld, Fakultät für Chemie, Physikalische Chemie I, Bielefeld, Germany

Svenja Einecke, Thomas Dreier, Universität Heidelberg, PCI, Heidelberg, Germany

Andreas Dreizler, Steffen Lindenmaier, Universität Stuttgart, Inst f Techn Verbrennung,
Stuttgart, Germany

Heinz Pitch, ITM-RWTH Aachen, Germany

Rune Kleiveland, NTNU, Inst f mekanik, termo- & fluiddynamik, Norway

Hakan Soyhan, Sakarya University, Istanbul, Turkey

Thomas Seeger, Martin Schenk, Joachim Jonuscheit, Lehrstuhl f Techn Thermodynamik,
Erlangen, Germany

Edward Blurock, Johannes Kepler Universität, Res Inst f Symbolic Computing, Linz, Austria

Richard Williams, University of Southampton, Great Britain

2.3 Academic Degrees Based on Work Carried out at the Divison during 1997-1998

Doctorates:

Nikola Georgiev, “Laser-based Techniques for Combustion Diagnostics”, 970521

Raymond Reinmann, “Theoretical and Experimental Studies of the Formation of Ionized Gases in Spark Ignition Engines”, 980515

Rolf Fritzon, “Resonant Laser Techniques for Combustion and Flow Diagnostics”, 980529

Hans Neij, “Development of Laser-Induced Fluorescence for Precombustion Diagnostics in Spark-Ignition Engines”, 981104

Licentiates:

Frederik Ossler, “Scattering of Light by Small Droplets in Sprays”, 970326

Sven-Inge Möller, “Detailed Numerical Simulation of Pulsating Combustion”, 971022

Joakim Bood, “Development of Dual-broadband Rotational CARS for Combustion Applications”, 981005

2.4 Seminars

Frederik Ossler, “Scattering of Light by Small Droplets in Sprays”, 970326

Derek Dunn Rankin, UCI, “Combustion Research”, 970421

Nikola Georgiev, “Laser-based Techniques for combustion Diagnostics”, 970521

Paul Ewart, Clarendon Lab, Univ of Oxford, “Non-Linear laser spectroscopy: from quantum mechanics to jet engines”, 970523

Sven-Inge Möller, “Detailed Numerical Simulation of Pulsating Combustion”, 971022

Edward Blurock, Johannes Kepler Univ, Linz, Austria, “Reaction: Modelling complex reaction mechanisms”, 971124


Heinz Pitsch, ITM-RWTH Aachen, Germany, “Flamelet Modeling of Non-Premixed Combustion Considering Differential Diffusion Effects”, 980122

Thomas Seeger, LTT Erlangen, Germany, “CARS spectroscopy at LTT Erlangen”, 980128

Raymond Reinmann, “Theoretical and Experimental Studies of the Formation of Ionized Gases in Spark Ignition Engines”, 980515

Rudolf Maly, Daimler-Benz AG, Stuttgart, Germany, “Combustion Research at Daimler-Benz”, 980515

Rolf Fritzon, “Resonant Laser Techniques for Combustion and Flow Diagnostics”, 980529


Mark Linne, Colorado School of Mines, Golden, USA, “Developments of Pico Second Pump/Probe Diagnostics”, 980925
Joakim Bood, “Development of Dual-broadband Rotational CARS for Combustion Applications”, 981005

Hans Neij, “Development of Laser-Induced Fluorescence for Precombustion Diagnostics in Spark-Ignition Engines”, 981104

Jean-Pierre Martin, Ecole Centrale, Paris, France, “Activities at the lab EM2c”, 981105

2.5 International Projects

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Human Capital & Mobility

HCM

Institutional fellowships

2.6 Budget

The Division’s budget for 1998 totalled 12.7 MSEK of which 80% came from external sources.

2.7 Undergraduate courses

Two undergraduate courses are given by the Combustion Physics Dep. Each course gives three credits, and a total of 180 credit points is required for the degree Master of science.

The course in Fundamental Combustion has been given since 1992/93, and normally 20 students are following the course. The course has during the last years been given by Per-Erik Bengtsson. The course literature has been produced by the course responsible and combined with the book by Griffiths and Barnard, Flame and Combustion, 3rd edition. The course consists of 16 lectures, two practicals (chemical kinetics, and flame velocity measurements), as well as demonstrations at different divisions and departments within the Lund Institute of Technology. The course is held for students on their fourth year of study in the disciplines of Engineering Physics, Engineering Chemistry, and Mechanical Engineering.

The course in Laser-based Combustion Diagnostics was held for the first time in the second study period of 1996/97. The course was given by Marcus Aldén, and the course literature was the book by Alan Eckbreth, Laser Diagnostics for Combustion Temperature and Species. The course consists of 14 lectures and two practicals (LIF and CARS). The course is given for students in Engineering Physics, and normally 10 students participate in the course. In addition, also some PhD students join the course.
3. LASER DIAGNOSTICS

3.1 Laser-induced Fluorescence

3.1.1 Fundamental Research

Picosecond Techniques

F. Ossler and T. Metz

The most important instruments in research involving picosecond (ps) resolution are a ps-laser system for excitation and a streak camera for detection. These are used in the study of fluorescence decays from flame excited species. In atmospheric flames the effective lifetimes lie around or below 1 ns, 100 to 1000 times shorter than the natural lifetimes. In some instances it is not necessary to use a streak camera. A fast photomultiplier tube and a fast oscilloscope are sufficient, if a deconvolution scheme is employed for the evaluation of the effective lifetime.

The ps-laser and streak camera have been used to demonstrate the possibility of performing two-dimensional measurements of the effective lifetime, without scanning procedures, in the measured volume. Operating the streak camera in the “focus mode” (no temporal deviation) and the “streak mode” (temporal deviation) two images are obtained. The latter contains the spatial and temporal information on one axis which can then be separated by employing a numeric algorithm (SAM = separation algorithm method) on the two images. This generates the two-dimensional distribution of the effective lifetime [Ossler, et al., 1996, Ossler, et al., 1998b]. In principle the method allows the lifetimes to be determined with a resolution set by the duration of the laser pulse and the resolution of the streak camera. In this way it is possible to determine lifetimes shorter than that expected of measurements with gated image intensifiers. The method was tested experimentally, as a proof of the principle, on different pieces of dyed and non-dyed paper. By this approach SAM could be compared with traditional time resolved lifetime evaluation along a line (TRM = time resolved method). As can be seen in Figure 3.1 the agreement between SAM and TRM was good. Finally, experiments were performed on two-dimensional objects and the corresponding images of the two-dimensional distribution of the effective lifetime were obtained with SAM, see Figure 3.2.

The present project is directed towards the characterization of different polycyclic aromatic hydrocarbons (PAH) in flames by means of spectral and temporal selection of the fluorescence emission. We have during the last year done picosecond laser induced, spectrally and temporally resolved fluorescence-emission measurements on the aromatic hydrocarbons: fluorene, naphthalene, anthracene and pyrene [Metz, et al., 1998, Ossler, et al., 1998a]. In order to study their temperature and oxygen-quenching behavior, the measurements were performed at atmospheric pressure conditions, first at temperatures between 150 °C and 950 °C and then at higher temperatures on naphthalene and pyrene seeded into a premixed flame.
The 266 nm wavelength radiation from a picosecond Nd:YAG was focused with a 1000 mm lens into a flow cell made of quartz. The cell was placed inside an oven and both had optical access for the incoming and outgoing laser beams as well as for the fluorescence emission, measured at 90°. By an optical arrangement consisting of UV-achromatic lenses mirrors and filters the fluorescence emission was focused onto a dual detection system, which included a spectrograph/OMA for the spectral analysis and a photomultiplier tube and a streak camera for the temporal analysis. Fluorescence quenching by oxygen was controlled by mixing the buffer gas (argon or nitrogen) with known amounts of air.

Results on fluorene, naphthalene, anthracene and pyrene show that the lifetimes decrease continuously with increasing temperature with about one order of magnitude. The spectral profiles are also sensitive to temperature: broadening and red shifts are observed for fluorene, naphthalene and anthracene. The spectrum of pyrene also broadens with increasing temperature but in difference to the other substances a new emission band appears on the UV side. Argon was in some cases used instead of nitrogen, e.g. for fluorene which showed relatively low stability at higher temperatures. The measurements showed also that it is possible to discriminate between fluorene and naphthalene in mixtures of the two by observing the decay of the fluorescence emission.

At lower temperatures (< 300 °C) all substances experienced quenching from oxygen in their fluorescence emission but at higher temperatures quenching became less clear. At even higher temperatures the gas mixtures reacted and no fluorescence emission was observed at all.

For the flame-related measurements the cell was replaced with a McKenna type burner and

![Figure 3.1: Fluorescence lifetimes of four pieces of dyed and non-dyed paper evaluated along a line using the two different techniques mentioned in the text.](image-url)
a mixture of naphthalene and nitrogen was seeded inside the flame. The laser radiation was focused at a height of ~7 mm above the orifice of the injector. By changing the stoichiometry of the premixed methane/air flame, different concentrations of oxygen were obtained around the injected gas. The PAH mixture burned like a candle shaped flame above the reaction zone of the premixed flame.

Time and spectrally resolved measurements for the case of no or very low oxygen concentration showed results, which could easily be extrapolated from the cell measurements. In fact by using the temperature dependence found for the lifetimes in the cell measurements,

**Figure 3.2:** An example for a two-dimensional object: A $5 \times 2$–matrix of dyed and non–dyed pieces of paper. The upper image (“focus-mode”) shows the intensity distribution, the lower one the lifetime distribution calculated using SAM.
the temperatures obtained from the lifetimes in the flame were not far (within 250 K) from the ones pre-calculated for the premixed flame without any seeding.

The project is at this moment proceeding with measurements on seeded flames.

Two-Photon Laser-Induced Fluorescence Detection of CO

_M. Richter and K. Nyholm_

Preliminary investigations of the behaviour of two-photon laser-induced fluorescence signals from carbon monoxide (CO) at elevated pressures have been performed earlier. The purpose of the present investigations was to study the applicability of two-photon excited LIF for one- and two-dimensional imaging of CO molecules in real combustion environments, such as internal combustion engines.

Carbon monoxide, like many other important combustion species, has its first electronic resonances in the vacuum ultraviolet region of the electromagnetic spectrum. Therefore, multiphoton excitation has to be utilised for the detection of this molecule, in order to overcome the problem of both generating and using vacuum UV light. We have applied laser-induced fluorescence by exciting the CO molecules from the ground state $\text{X}^1\Sigma^+$ to the excited state $\text{B}^1\Sigma^+$ using two photons at 230 nm and by observing the fluorescence from the excited level to the lower lying $\text{A}^1\Pi$ level. The fluorescence was detected in the spectral range between 451 nm ($v'=0$, $v''=0$) and 662 nm (0,5). Spectrally and spatially (in one
dimension) resolved measurements were carried out in a cell, as well as, in a flame envi-
ronment. The focal region of the laser beam focused to a line was imaged into a spectrom-
eter equipped with a CCD camera.

The new contributions to this field of research are experiments performed under similar
conditions as in earlier investigations, but with higher laser energies. The reason for this is
to study the power dependence and the effects of ionisation of the CO molecules. Nor-
mally, the fluorescence signal in two-photon excitation processes depends quadratically on
the incident laser intensity. However, as the level of ionisation increases the power depen-
dence moves towards the linear regime. A YAG-pumped OPO laser was used to generate
approximately 12 mJ of excitation light at 230 nm. This should be compared with the 3-4
mJ that were available in previous experiments.

The two-photon fluorescence signal from CO was studied as a function of the laser fluence
and gas pressure. Measurements in a cell filled with different buffer gases were carried out
at pressures from 0.1 up to 8 bars. In the flame investigations, the behaviour of the spectral
interference from nonresonantly excited C₂ molecules, produced by photodecomposition
of fuel and fuel fragments were studied at various CH₄/air–flame stoichiometries and laser
intensities.

Two-Photon Laser-Induced Fluorescence for Detection of NO

C. Kaminski, Ö. Andersson, and J. Engström

When detecting nitric oxide (NO) by LIF, a common source of interfering fluorescence is
hot oxygen, which fluoresces effectively upon illumination in the Schumann-Runge bands,
as well as Stokes shifted fluorescence from large hydrocarbons. In the present study it was
investigated whether these difficulties could be overcome by employing a two photon ex-
citation scheme for NO.

Using sufficiently intense laser radiation, a molecule may absorb two photons if the sum of
their energies matches an electronic resonance. Intuitively, one can imagine that the first
photon excites a ‘virtual’ level, from which the second photon completes the transition to
a real level. The effective excitation energy corresponds to the sum of the frequencies of
the two photons.

Since two photons are involved in the excitation, the signal scales as the laser intensity
squared. Furthermore, due to different electric dipole selection rules two-photon spectra of
NO contain O and S-branches since, in contrast to one photon selection rules, transitions
with ΔJ = ±2 are allowed. An inherent weakness, however, is the low signal levels ob-
tained, due to the low two-photon absorption cross-section. In practice therefore, a large
photon flux is required to optimise the signal to noise ratio. However, the spectral profile is
dependent on the electric field strength and a characteristic feature of the two-photon spec-
trum of the g-bands of NO is a large a.c. Stark broadening coefficient. The Stark broadened
lineshapes are asymmetrical, which makes the interpretation of spectra of NO at atmo-
spheric pressure or above difficult owing to significant spectral overlap between individual transitions.

Initial lineshape experiments were performed in a low pressure cell containing NO. Two-photon LIF signals from the cell were detected by a photomultiplier tube. To determine the detection sensitivity of the technique in more realistic environments, experiments were performed in well-characterised $\text{N}_2\text{O}/\text{H}_2$ flames that could be seeded with NO. The signal’s dependence upon various flame parameters was measured, and the obtained results were compared with detailed flame calculations to obtain estimates of the sensitivity of the technique. Saturation checks could be performed using a variable beam attenuator. Pulse energies were monitored on a fast photodiode.

The cell experiments revealed the onset of significant asymmetric broadening of the type discussed above already at intermediate laser pulse energies (~3 mJ). Excitation scans were also performed in a laminar, premixed $\text{N}_2\text{O}/\text{H}_2$ flame to determine the location of the strongest line for excitation. Because of poor signal to noise ratios (SNR) on nearly isolated transitions within the $\gamma$-bands of NO, the bandhead near 452 nm was chosen for excitation since it provided the strongest signal levels and thus the best potential for imaging applications.

The LIF signal level was measured as function of laser power, and a power dependence of approximately two was found at all pulse intensities employed. This suggests the fluorescence was not saturated which is explained by the low transition probabilities of two-photon techniques. Despite of these findings, asymmetric Stark shifts were apparent at pulse energies higher than 3 mJ.

To test the sensitivity of the technique, measurements were performed under various flame operating conditions and the results compared to detailed calculations of NO chemistry in the flame. Two photon NO-LIF signals were measured as a function of flame stoichiometry, flame height, and amounts of NO added to the gas mixture. All measurements were point measurements obtained with a photomultiplier tube. The calculations were based on a one-dimensional application of the GRI-mechanism employing full chemistry and the theoretical curves fitted to the measured concentration data. These measurements were referenced to spatially resolved measurements employing an intensified CCD camera from which the detection sensitivity of the present technique could be estimated. Concentrations of naturally occurring NO in the flame could be successfully imaged along a line with single shot SNR’s exceeding 10:1. From such measurements and the flame calculations it could be estimated that the absolute detection limits of the technique were of the order of 1000 ppm of NO at atmospheric pressure (single shot). [Kaminski et al., 1997]
Investigations of Polarisation Spectroscopy for Combustion Diagnostics

K. Nyholm, J. Walewski, B. Löfstedt, A. Dreizler, T. Dreier and T. Karlsson

In PS the signal is generated as a laser beam, so that the necessary solid angle for the detection channel is typically three orders of magnitude smaller than for LIF. Moreover, spatial filtering in the focus plane of an imaging lens makes it possible to suppress the non-coherent background even more. Therefore PS has the potential of a measurement technique for minor species in environment with high scattering, e.g. sooty flames. Earlier it was demonstrated, that the PS signal of OH in a diffusion flame after filtering through an aperture does not show any background from the non-resonant Rayleigh scattering at soot particles. This interesting feature was reinvestigated and this time compared with LIF. For this purpose PS and LIF signals from OH radicals at 308 nm were recorded in a welding torch burner for different heights above the nozzle and different ratios of the premixed propane/oxygen mixture. The signals were recorded along a line to make spatial distinction of the resonant signal from OH and non-resonant scattering possible [Dreizler et.al.,1998]. For the case of a pure diffusion flame the OH signal to scattering ratio was at least higher than 10:1 for PS, while it was less than 1:2 for LIF signals at the same height. The origin of the background in the PS signals was identified as non-resonant Rayleigh scattering from the strong pump beam at soot particles, while the probe beam was not influenced by the soot particles. This opens up the possibility to get even higher background suppression by additional spatial filtering of the probe beam. With this method it

Figure 3.4: PS signal from CH$_4$ at 3017 cm$^{-1}$ while varying the pressure of a buffer gas (N$_2$). Pressure dependence (buffer gas) of Q(4) and Q(6) at 96 mTorr methane.
may therefore be possible to measure in even more sooty environments [J. Walewski et. al., 1998].

Although PS has been shown to be a powerful technique for combustion diagnostic of both species concentration and temperature, little progress has been made during the last years in applying PS for measurements in real combustion processes. The reasons for this stagnation are mainly two:
1. Until now no applicable theory for PS in the used conditions (high pulse power, high temperature, high pressure) is available.
2. The influence of the gas pressure on the signal may be very strong and can lead to the extinction of the signal at atmospheric pressures or higher. This has been shown in a joint measurement at Sandia National Laboratories in cooperation with Dave Lakeshaw. In these experiments, CH$_4$ in a gas cell was excited at 3017 cm$^{-1}$. It was not possible to get any PS signal at atmospheric pressure, first at pressures below 200 mbar a PS signal was observed (see Figure 3.4). This stands in strong contrast to measurements of other molecular species in flames, e.g. OH and C$_2$, for which a PS signal is easily received at atmospheric pressure.

To investigate this phenomenon in order to identify processes causing the strong dependence, measurements were carried out at our laboratories. PS on the OH radical in the flame of a low pressure burner was investigated for this purpose. The aim of the measurements is to compare LIF and PS signal and spectrum for different pressures and different heights above the burner. These measurements are not completed, yet [Dreizler et.al., 1998].

3.1.2 Applied Research

Instantaneous 2D Temperature Measurements in Combustion by Atomic Laser-Induced Fluorescence

C. Kaminski and J. Engström

Data on temperature field distributions of turbulent combustion processes can provide important input for the improvement of theoretical models and the optimisation of practical combustion devices. Different laser diagnostic techniques have been applied in practical combustion to measure temperature field distributions. The present group has developed a new laser technique for measurement of temperatures in combustion processes [Kaminski et al., 1997]. The technique is based on two line atomic fluorescence (TLAF) from seeded atoms. Time resolved, two-dimensional thermometry in turbulent combustion has for the very first time been demonstrated with TLAF [Kaminski et al., 1998]. The basic principle of TLAF is that suitable metal atoms having two optically accessible, temperature sensitive energy states, are seeded into the combustion environment. Depending on the temperature the atomic energy levels are populated according to a Boltzmann distribution under conditions of local thermodynamic equilibrium. Two time delayed laser pulses at wavelengths $\lambda_{02}$ and $\lambda_{12}$ are used to probe the ground state 0 and first excited state 1 of the atoms by driving transitions to a common upper state 2. Upon excitation, level 2 will
fluoresce back to the two available lower states 0 and 1 yielding LIF signals $F_{20}$ and $F_{21}$, respectively.

Two filtered detectors collect the LIF signals corresponding to $F_{20}$ and $F_{21}$. In thermal equilibrium the ratio $F_{21}/F_{20}$ is proportional to the temperature $T$ of the system. It can be shown that

$$T = \frac{\varepsilon_{01} / k_B}{4\ln \frac{\lambda_{21}}{\lambda_{20}} + \ln \frac{I_{12}}{I_{02}} + \ln \frac{F_{21}}{F_{20}} + C} \quad (3.1)$$

where $\varepsilon_{01}$ is the energy difference between the two lower states involved (in J), $k_B$ is Boltzmann’s constant, $\lambda_{2i}$ are the fluorescence wavelengths, and $I_{2i}$ and $F_{2i}$ ($i=0,1$) are the laser and the fluorescence signal intensities, respectively. $C$ is a system dependent calibration constant. The equation is valid under the assumption of a linear relationship between laser intensity and LIF signal. Since the oscillator strengths of atomic seeder species can be of the order of unity the effects of saturation and fluorescence trapping have to be carefully checked for in a given measurement situation.

The technique has been successfully applied to a near production automotive engine running on isooctane fuel and a pulse combustor running on methane gas. The temperature sensitivity of the technique allows measurements of burned gas covering a temperature range from 700 to 2800 K. A precision of 14 % on a single shot temperature distribution has been obtained. A figure that could be substantially improved if online intensity profile measurements of the laser sources is performed. The technique does not suffer from quenching and requires only very low pulse power densities for adequate SNR’s to be obtained. In Figure 3.5 the temperature distribution and the flame propagation from the SI engine application are shown. The flame is seen to develop to the right hand side of the viewing field, which corresponds to the position of the exhaust gas valves.

Furthermore, the technique has shown a potential for two-dimensional temperature measurement in sooting environments [Nygren, 1998]. The particular properties of the present technique (low laser intensities required, excitation wavelength in the visible, distant from major spectral interference) are advantageous in a sooting environment.

\[ \text{Figure 3.5: Excitation schemes for TLAF using indium.} \]
Laser Spectroscopic Investigation of NO Formation in a Realistic SI Engine

F. Hildenbrand, C. Schulz V. Sick, G. Josefsson, I. Magnusson, and Ö. Andersson

The distribution of nitric oxide (NO) was measured in a spark ignition (SI) engine fueled with a stoichiometric iso-octane/air mixture. Different engine operating conditions were investigated, and accurate NO concentrations were obtained from essentially the whole cylinder for crank angle ranges from ignition to the mid expansion stroke. Laser-induced fluorescence (LIF) was used to measure the two-dimensional NO concentration distributions, utilizing a KrF excimer laser to excite the NO A-X (0,2) bandhead. Results were achieved with high temporal and spatial resolution. The NO images were also used to evaluate the flame development. Both the mean and the variance of a combustion progress variable could be deduced. The measurement object was an optically accessible one-cylinder version of the VOLVO N1P engine.

The fluorescence intensity is given by

\[ I_{\text{LIF}} = I_{\text{laser}} \cdot N(p,T) \cdot V \cdot f_B \cdot B_{ki} \cdot g_\lambda(p,T) \cdot \frac{\tau_{\text{eff}}}{\tau_{\text{rad}}} \cdot \frac{\Omega}{4\pi} \cdot \varepsilon, \]

(3.2)

where \( I_{\text{laser}} \) is the laser intensity, \( N(p,T) \) is the NO number density in the measurement volume, \( V \), and \( f_B \) is the fraction of NO molecules residing in the lower level of the transition. \( B_{ki} \) is the Einstein coefficient for spontaneous emission and \( g_\lambda \) measures the optical overlap between the laser and the absorption line. The ratio \( \frac{\tau_{\text{eff}}}{\tau_{\text{rad}}} \) represents the fluorescence quantum yield, here mainly depending on the quenching rate. The last two factors represent the detection solid angle and signal losses in the detection system. In order to

Figure 3.6: Temperature distribution and flame propagation obtained by imaging TLAF in an SI-engine.
evaluate $N(p,T)$, all other factors have to be known. The pressure and temperature varies significantly during the engine cycle. Thus is $g_λ$ critical, since NO suffers from extreme pressure broadening and shift. The quenching rate is also critical and was determined from calculated equilibrium concentrations of the most important quenching species.

It can be clearly seen in Figure 3.7 that the NO formation starts near the spark plug in the middle of the combustion chamber. In early stages of the combustion the flame and thus the NO concentration field is shifted towards the exhaust valves due the tumble flow. A steep increase in NO concentration can be seen as long as pressure and hence temperature
is rising. After 10° after top dead center, the NO mole fraction remains more or less constant whereas the number density decreases due to the decreasing total pressure. [Hildenbrand et al., 1998]

Applications of a multiple-pulse Nd:YAG laser and framing camera system for ultrafast visualisation of combustion processes

J. Hult and C.F. Kaminski

A new laser and detector system for high speed imaging of turbulent combustion phenomena has been set up at the Division of Combustion Physics. The system is capable of delivering up to eight laser pulses in rapid succession and to capture two dimensional images of corresponding events. The system can be used for a number of established spectroscopic methods for combustion diagnostics, but the high repetition rate of the system extends these methods to the time regime where highly turbulent or other dynamic phenomena can be resolved. [Kaminski et al., 1999b]

The laser source consists of four individual Nd:YAG lasers built into one unit. Each individual laser consists of an oscillator and a single amplifier. By switching each Pockels cell twice during the flash lamp pulse the lasers can be operated in double pulse (DPO) mode, thus allowing a burst of a maximum of 8 pulses to be emitted in series from all cavities. The four lasers are individually controlled and the individual pulses can be synchronised with respect to each other or to arbitrary external events. A special beam combining scheme is used for combining the output from the four cavities into a single beam.

Figure 3.8: Spark ignited turbulent flame development in a combustion cell equipped with high speed ventilators to generate controlled amounts of turbulence. The images in the upper row are OH PLIF images, the bright areas indicate burned gases in a cut through the centre of the flame. The images in the lower row are flame emission images recorded simultaneously, these are integrated line of sight images indicating the maximum extent of the flame.
For detection an ultra fast framing camera, (Imacon 468, Hadland Photonics) is used. The camera consists of 8 independent CCD detectors. The image is split into 8 identical images by an 8 facet pyramid beam splitter and the resulting images are relayed to the individual CCDs. The individual CCDs have their own intensifiers and are independently gateable, thus giving full timing control. An option is provided to attach an extra 3 stage intensifier to the Imacon. This intensifier increases the detection sensitivity of the camera and also extends its operation into the UV spectral region.

A PLIF spectrometer for time resolved studies of radical concentration fields in turbulent combustion environments has been built up, it is based on a standard dye laser pumped by the multiple pulse Nd:YAG laser system.

Time resolved studies of flame propagation utilising PLIF of the OH radical as a flame marker have been performed in a combustion cell. The cell was equipped with fans to generate turbulence of variable intensity. This experiment offered the possibility to study the interaction of turbulent flow phenomena and flame chemistry in real time for the first time. Studies of the flame kernel development were performed for different degrees of turbulence and for different gas mixtures. Flame emission images were recorded simultaneously with the PLIF images, thus both line of sight images and 2D-cuts through the

Figure 3.9: OH PLIF images of a turbulent diffusion flame, the inner flow is air and the outer flow is methane. Time is increasing for pictures going from left to right and from top to bottom, respectively. The time between consecutive images corresponds to 125 µs.
flame were recorded. The results from these measurements will be compared to the results
from both DNS and PDF modelling. The ignition system of the cell was designed to yield
highly reproducible ignition events, which was verified by studying the evolution of lami-
nar flames ignited by the system. Standard deviations of the flame radius at a certain time
after ignitions were found to be about 1%. The results from the PLIF data were directly
compared to detailed numerical simulations modelling spark ignition of laminar premixed
flames. The OH PLIF studies in the combustion cell were performed as part of an LLC
(Lund Laser Centre) project together with A. Dreizler and S. Lindenmaier, Universität
Stuttgart.

Fuel visualisation by PLIF of acetone seeded into the fuel gas has also been attempted in a
combustion cell. OH and acetone as flame front markers have been compared by taking OH
and acetone PLIF images when running the cell under identical conditions. The two tech-
niques were shown to mark different flame front regions, this effect occurs because zones
of acetone pyrolysis and OH radical generation do not coincide. The acetone PLIF studies
in the combustion cell were performed as part of a LLC project together with R. Williams,
University of Southampton.

Time resolved measurements of radical concentration fields have also been performed in a
turbulent diffusion flame. The OH radical concentration in the flame was measured using
the same PLIF spectrometer as for the cell studies. Time resolved images series of the
turbulent flame were taken for different Reynolds number flows and at different distances
from the burner nozzle. Visualisation of the flow without combustion has also been per-
formed by two different techniques. In the first technique one flow was seeded with ac-
etone and visualised by PLIF of acetone, in the second technique the same flow was seeded
with smoke particles of micro meter size and visualised by Mie scattering from these smoke
particles. By comparing the images taken with and without combustion the effects of com-
busion on the turbulent flow field can be determined. [Kaminski et al., 1999]

Applications of Laser/Optical Diagnostics for the Characterization of Practical
Low-Emission Gasturbine Combustors

C. Löfström, J. Engström, M. Richter, C. F. Kaminski, K. Nyholm, J. Hult, J. Nygren, and
M. Karlsson

Measurements were made to investigate an aircraft combustor in collaboration with Volvo
Aero Corporation. A combustor with a new concept in order to meet high efficiency com-
bined with ultra-low emission has been constructed by VOLVO Aero Corp. The construc-
tion is called a lean premixed prevaporized (LPP) combustor. The measurements were
made with the aim to both characterise LPP combustors using mature techniques and fea-
sibility studies of novel techniques and their applicability in harsh industrial environments.
Four laser diagnostics experiments were performed: Planar Mie/LIF experiments to estab-
lish the degree of vaporisation, OH PLIF-imaging, two- line atomic fluorescence (TLAF)
to obtain 2-dimensional temperature distributions, and detection of CO using two- photon
LIF. In order to investigate flame fluctuations an ultrafast framing camera was employed to
record spontaneous emission at rates up to 50 MHz. The preliminary results were pre-
sented at the 27th International Symposium on Combustion [Löfström et al., 1998]

The LPP combustor was operated on JET A as fuel mixed with air whose temperature was preheated to ~600K at a pressure of 7 bar. The principal features and the measurement region of the duct is shown in Figure 3.10. A pilot flame is situated in the centre of the duct where measurements were performed and the premixed air/fuel is supplied through the exterior duct. Two windows just behind the duct along the chamber walls provide horizontal optical access for the laser sheet. From a third window, located in the bottom of the chamber, the fluorescence was collected.

The main experiment was to investigate the degree of vaporisation using Mie scattering from droplets simultaneous with LIF imaging of the fuel distribution. Measurements were made both with and without combustion. The Mie scattered light mark areas where fuel is in the liquid phase. The fluorescence light comes both from vapour and liquid phases. To get an image of the degree of vaporisation, both Mie and fluorescence images were combined. Assuming that regions with Mie signals above a selected cut-off value corre-

![Figure 3.10: Principal features of the duct and measurement regions. The laser sheet passes through the windows and the lean areas close to the windows are clearly visible.](image1)

![Figure 3.11: Vapour distribution determined by a combination of LIF and MIE scattering.](image2)
spond to the liquid phase created the liquid image. Corresponding areas in the fluorescence images were marked with black colour in the vapour images. The resulting image then shows how the vapour is distributed around the liquid (Figure 3.11).

The TLAF experiment was performed with the aim to measure a temperature distribution and to investigate the potential of the technique in the LLP environment. The TLAF technique is discussed in more detail earlier in this section. In this very lean combustor the signal levels obtained were relatively low. One possibly reason is rapid oxidisation of the atomised indium by the preheated air and corresponding signal loss, for further discussion see [Engström, 1999]. A mean temperature distribution referenced to a theoretically obtained mean temperature based on 20 single shot TLAF image pairs is shown in Figure 3.12.

The potential for CO detection was also investigated by excitation via a two-photon excitation scheme near 230 nm followed by fluorescence emission in the visible. Two-photon excitation schemes can be utilised to overcome problems such as strong absorption by ambient air or generation of laser light in the far UV spectral region.

**Fuel visualisation in a SI-Engine Using Laser Induced Fluorescence Signals Collected Through an Endoscopic Detection System**

*M. Richter and B. Axelsson*

The basic idea when performing fuel visualisation in a SI-engine is to run the engine on a one-component fuel such as isoctane mixed with a fluorescent tracer e.g. 3-pentanone or acetone. A laser beam formed to a thin sheet then excites the tracer. The fluorescence from the tracer is then detected from an angle perpendicular to the laser sheet. This result in a two-dimensional image showing the relative fuel, i.e. tracer, concentration across the area covered by the excitation light.

LIF and other laser techniques proclaim optical access to the combustion chamber. The most common approach to establish that is to use a cylinder liner partly made of glass and, when possible, small windows in the upper part of the combustion chamber, i.e. in the
cylinder head. In order to make perpendicular detection possible a window is placed in the piston. A mirror is placed in the block under the see-through piston to guide the signal to a detector. By using this design two-dimensional images can be recorded by a CCD-detector equipped with conventional camera lenses.

This so called Boditch-design works fine for most applications but problems arise when engines with advanced piston crown designs are to be studied. Due to the complex shape of piston crowns, windows can not be placed in an arbitrary manner and that inevitably limits the field of view when through the piston detection is used. In addition, it is not uncommon that a curved surface on the piston crown has to be replaced by the flat surface of an inserted window, e.g. when a window is placed in the bottom of a piston bowl. Such changes might introduce undesired changes to the flow field and hence give rise to unrealistic measure conditions.

An endoscope on the other hand can, independently of piston design, be inserted somewhere in the cylinder head. Thus measurements can be performed under more realistic conditions (load, pressure, temperature) and with the blueprinted piston design. This is an advantage when engines where the piston shape is critical for the motion of the charge are studied.

An endoscopic detection system has been used for LIF visualisation of both gaseous and liquid fluids in a DISI-engine. The use of an endoscope made it possible to monitor parts of the combustion chamber that could not be accessed through the piston with conventional optics. Brief investigations of the signal collection efficiency have been performed on the

Figure 3.13: Schematic view of the optical access system to the combustion chamber (left), and a photograph of the piston top as viewed from above (right).
endoscopic system as well as on a system based upon conventional optics. The technique shows promising results and the use of endoscopic detection systems should be considered as a compliment to using advance design quarts piston crowns for conventional, through the piston detection. [Richter et al., 1998a]

Investigation of the Fuel Distribution and the In-cylinder Flow Field in a Stratified Charge Engine Using Laser Techniques and Comparison with CFD-Modelling

M. Richter and B. Axelsson

The combustion characteristics of a stratified charge SI-engine are to a large extent determined by the in-cylinder flow field. In order to achieve a stable combustion it is necessary to have control over the stratification process, i.e. to place the fuel in such manner that there is an ignitable mixture around the spark plug at the time of ignition. The motion of the evaporated fuel is controlled by the induction flow of fresh gases in combination with the properties of the initial fuel spray. These two flows interact with each other, and together they produce a non-trivial flow pattern.

In a project, aiming at increased understanding of the large-scale stratification, together with VOLVO Car Corporation, different laser techniques has been applied to a Direct Injection Spark Ignition (DISI)-Engine with optical access. Laser Induced Fluorescence (LIF) visualisation of the fuel distribution has been performed using an endoscopic detection system. Due to the complex piston crown geometry it was not possible to monitor the critical area around the sparkplug with conventional, through the piston, detection. Therefore, an endoscope inserted in the spark plug hole was used. This approach gave an unrestricted view over the desired area. In addition, the in-cylinder flow fields have been monitored by Particle Image Velocimetry (PIV). Measurements were performed through cylinder and piston so that both horizontal an vertical flows could be monitored.

The results from the LIF and the PIV measurements have been compared with CFD-modelling at Volvo. The validation was made at part load when the engine was operating in stratified mode, i.e. late injection during the compression phase. Qualitative agreement was found between the calculated and measured fuel distribution around the spark plug prior to ignition. Also, the PIV measurements showed a good agreement with the flow fields obtained by CFD-modelling. In addition, the transportation properties of the fuel distribution that was monitored by LIF could to a great extent be explained by the results from the PIV measurement and the CFD-modelling. All three techniques showed promising agreements with each other and the measured properties could be used to further increase the accuracy of the CFD-modelling. The close collaboration and comparison between different techniques increased the understanding of the processes going on in the combustion chamber. [Richter et al., 1999]
Figure 3.14. Fuel distribution monitored by LIF (above) and calculated (CFD) fuel distribution (below)
3.2 Other Techniques

Rotational Coherent anti-Stokes Raman Spectroscopy

J. Bood, C. Brackmann, and P.-E. Bengtsson

The temperature is an important parameter in the characterization of combustion processes, and the temperature distribution has a strong connection to the emission of pollutants and the efficiency of such processes. It is a difficult task to perform accurate in-situ temperature measurements in real practical systems, and the development of laser techniques has been very important for the progress in this area. The variant of CARS named rotational CARS, which stands for rotational coherent anti-Stokes Raman spectroscopy, has been further developed and investigated in different projects.

In Figure 3.15, a typical experimental set-up is shown for dual-broadband rotational CARS. In DB-RCARS, three laser beams are focussed to a common intersection point from which a signal is generated if molecules with suitable resonances are present. The signal is generated as a laserlike beam and directed to a spectrograph and a detector. The spectrally resolved signal is analysed by fitting its shape to a library of theoretically calculated spectra at different temperatures. The temperature is evaluated from an interpolation between the theoretical spectra. An example of an experimental rotational CARS spectrum is shown in Figure 3.16, together with the difference spectrum between the experimental spectrum and the best-fit theoretical spectrum. The spectrum was recorded from measurements prior to ignition in a spark-ignition engine at a pressure of 19.7 bar, and the evaluated temperature was 838 K [Bood et al., 1997]. Both oxygen and nitrogen lines are present in the spectrum. The phenomenon of engine knock in spark-ignition engines is studied in a collaboration project consisting of the following sub-projects: evaluation of knock detection methods, modelling of flow and flame propagation, modelling of chemical kinetics, thermal analysis and heat transfer, and our contribution on laser diagnostic temperature measurements using DB-RCARS [Bood et al., 1997].

Figure 3.15: Experimental set-up for dual-broadband rotational CARS.  Figure 3.16: Example of a rotational CARS spectrum.
A single-cylinder spark-ignition engine was modified for the experiments. The spacer between the cylinder liner and the cylinder head is shown in Figure 3.17. The engine is designed to give zero swirl and is equipped with two spark-plugs to provide as plane flame front as possible and thereby a well-defined end-gas region close to the wall. This anticipated end-gas region is probed using the DB-RCARS technique. In order to be able to perform CARS measurements close to the wall, a vertical planar BOXCARS configuration was used (see Figure 3.17). To further decrease the shortest possible distance between the probe volume and the wall, the wall is not completely plane but slightly wedged. The focusing and recollimating lenses were mounted on two translators, making it possible to accurately translate the laser beams, and consequently the probe volume, orthogonally to the wall inside the combustion chamber. Since the measurements were performed in an engine with optical access through two quartz windows, and in a point close to a surface, scattered stray light from the Nd:YAG laser (532 nm) had to be suppressed using an optical notch filter.

During the measurements the engine was running at 1200 rpm, triggering the Nd:YAG laser. The compression ratio of the engine was 5.5:1 and the fuel was a mixture consisting of 60\% iso-octane and 40\% n-heptane (RON 60). The ignition time was set at –27 crank angle degrees (CAD), and measurements were performed at 17 different timings between –42 and +26 CAD relative to top dead center (TDC). At each time 100 single-shot spectra were recorded and evaluated individually. A comparison between the measured and calculated temperatures is shown in Figure 3.18, for measurements where the probe volume was located at a distance of 3.0 mm from the wall. For CAD:s from +16 and later the number of spectra possible to evaluate decreases rapidly. This behaviour reflects that an increasing number of cycles cannot be evaluated because of autoignition in the end-gas.
The DB-RCARS technique was used to measure cycle-resolved temperatures prior to ignition in the end-gas of the engine. The accuracy of the evaluated temperature in each single-shot is estimated to be better than 30 K at around 800 K.

The European EUREKA Gas Turbine program AGATA (Advanced Gas Turbine for Automobiles) was started in 1993 with the aim to develop three critical components for a 60 kW turbogenerator in a hybrid electrical vehicle, and one of these was a catalytic combustor. Simultaneous temperature and relative oxygen concentration measurements were performed after this catalytic combustor [Bood et al., 1999]. The aim of the project was to investigate

**Figure 3.19:** Catalytic combustor set-up.

**Figure 3.20:** Evaluated temperature and relative oxygen concentration profiles using DB-RCARS.
the performance of the catalytic combustor for different running conditions in a full-scale pilot study.

The catalytic combustor is illustrated in the drawing presented in Figure 3.19, and the CARS section is the part of the system where the optical measurements were performed. Since the probe volume had to be moveable over a distance of 100 mm the focusing and recollimating lenses were mounted on two translators remotely controlled by a PC.

DB-RCARS measurements were generally performed in eight points along a line in the measurement section. At each point three accumulated spectra, each consisting of 300 single-shot spectra, were recorded. Measurements were performed at different running conditions corresponding to different gas turbine power levels. The measured temperature and oxygen concentration profiles for one condition is shown in Figure 3.20, and the position 0 mm corresponds to a position right in the middle between the two windows. A correlation between the measured temperatures and oxygen concentrations can be observed: high temperatures corresponds to low oxygen concentrations and vice versa, reflecting the varying degree of combustion efficiency.

In the presented measurements, the conditions in the combustor were considered to be constant during the measurement period. This meant that a large number of single-shot spectra could be accumulated to an averaged spectrum. Such averaging leads to a higher accuracy in the measurement and the temperature is estimated to be evaluated with an uncertainty less than 20 K at 1500 K.

A common experimental problem using rotational CARS, especially in applied situations, is undesired spectral interference due to stray light from the primary laser beams. An atomic filter in the form of a sodium-seeded flame has been demonstrated for suppression of stray light originating from the narrowband laser radiation in DB-RCARS experiments [Bood et al., 1998]. The filter consisted of an acetylene/air flame on a slot burner seeded by sodium chloride. By the use of an additional dye laser tuned to 589.0 nm as the narrowband source the sodium filter effectively absorbs stray light at the sodium D2-line (589.0 nm), without any signal loss or spectral distortion.

In most DB-RCARS experiments, the temperature is evaluated by spectral fitting of experimental spectra to a library of theoretical spectra. There is a lot of work in constructing theoretical libraries from molecular data and experimental parameters. Also, the fitting procedure may be time-consuming in the evaluation of a large number of single-shot spectra. Quick-fit methods may be a solution to the problem. A quick-fit method based on the ratio of the intensities in different spectral regions was developed and evaluated regarding accuracy and precision [Brackmann, 1998]. The results were promising and the characteristics of the method will be further investigated in the near future.
Soot Diagnostics

B. Axelsson, R. Collin, and P.-E. Bengtsson

In fires, soot is the main source of heat radiation and thus an important factor in fire spread. It is heated soot particles that makes the log fire a useful heat source and the candle to a convenient source of light. However, in many technical combustion systems soot is a major pollutant. As particulate and NOx emission regulations are getting stricter, increased attention has to be devoted to the understanding of soot formation and oxidation. By this, possibilities to reduce soot emission in technical combustors, such as Diesel engines, increases. One reason to the lack of understanding is the absence of suitable diagnostic techniques for probing combustion processes. Most of the techniques used have severe limitations concerning temporal and spatial resolution. Laser-based techniques, however, have the potential to provide better temporally and spatially resolved measurements of the soot distribution.

Laser-induced incandescence (LII) is today a well-established technique to visualize soot volume fraction. The technique is based on the heating of particles by absorption of incident laser radiation. The soot particles are heated to temperatures of 4000 to 5000 K and the resulting blackbody radiation is detected. The induced incandescence signal has shown to be approximately proportional to the soot volume fraction in the probe volume. A particle with higher temperature will radiate stronger at shorter wavelengths according to Wien’s displacement law. By detection close to 400 nm, an efficient reduction of the background emission is achieved and the soot can then be detected.

Introductory measurements of the soot volume fraction have been performed with the LII-technique in a premixed one-dimensional sooting flame. The LII-technique was calibrated with an extinction measurement in order to make quantitative measurements. Two-dimensional LII measurements of the soot volume fraction with a laser sheet were compared with

![Figure 3.21: Decay of the LII signal as a function of particle size.](image)

Intensity / a.u. vs. t / ns for particle sizes 10nm, 15nm, 20nm, 30nm
two approaches of the one-dimensional measurements with a focused laser beam. The measurements from the two LII-techniques were also compared with measurements performed with the extinction technique. The outcome of this introductory investigation was that the results from the different measurements seem to match each other for lower, undisturbed heights in the flame. This work was presented in a diploma work [Collin, 1998].

In a cooperation with the department of fire and safety engineering, the LII technique was applied to measure soot volume fraction. The measured values will be used to validate models of the formation of soot in turbulent diffusion flames. The cooperation took place within the CECOST soot project. The soot project is the first CECOST project and it started in 1997. For more information about CECOST see: URL:http://www.fysik.lth.se/cecost/.

Preparatory work to measure soot particle size has been performed and the actual measurements have been initiated. The technique to be used is time resolved laser-induced incandescence (TIRE-LII). Since larger particles cool slower the size can be determined by observing the decay of the LII-signal.

The preparatory work consisted of the characterization of flames burning on a porous-plug burner. The soot volume fraction and particle size was measured with extinction/scattering. The characterized flame will be used for calibration purposes. The picture below shows the soot volume fraction and the particle size as a function of height above burner (fuel equivalence ratio, $\phi=2.3$).

**Figure 3.22:** Soot volume fraction and particle size as a function of height above burner ($\phi=2.3$).
Real-time calibration of LIF air/fuel-ratio measurements in combustion environments using in situ Raman scattering

M. Richter and B. Axelsson

The increasing need to reduce pollution and improve fuel economy in the field of transportation has led to the concept of ultra lean-burn SI-engines. In order to provide an ignition and a mixture supporting flame kernel growth at the spark plug in such an engine, a well-defined inhomogeneous charge distribution is required. The design goal is to provide a near stoichiometric or slightly rich gas mixture around the spark plug at the time of ignition, while the total charge shall remain lean. This of course complicates the process of mixture preparation, since besides achieving a good local mixture, it is also necessary to have control over the large-scale stratification.

Relative species distributions can be monitored two-dimensionally by laser-induced fluorescence (LIF) imaging techniques. This is today a widely spread technique, in the development of combustion engines, that provides the designers with qualitative information on the fuel distribution. Due to quenching, i.e. de-excitation caused by collisions, LIF measurements do normally at best only provide relative information on the fuel distribution. Therefore a calibration method is required to achieve absolute air/fuel data.

A laser-based technique for recording such absolute air/fuel-ratio maps in a single laser pulse in combustion devices, for instance prior to ignition in internal combustion engines, is under development. The method is based on an in situ pulse-to-pulse calibration of two-dimensional laser-induced fluorescence (LIF) images of relative fuel distributions by a point Raman measurement. A single excimer laser pulse is used to quasi-simultaneously

Figure 3.23: The LIF signal (above) shows the 2D fuel distribution. The Raman signal (below) is used to determine the air/fuel-ratio in a specified area.
detect planar laser-induced fluorescence from a fuel tracer as well as spectrally resolved Raman scattering from oxygen and fuel in one point inside the LIF image plane. By determining the air/fuel-ratio in this particular point from the measured Raman signals, the whole LIF image can be scaled, and quantitative two-dimensional air/fuel-ratio data obtained. In the method, it is crucial to achieve a Raman signal that can be spectrally separated from the much stronger LIF signal. Therefore the behaviour of the Raman signals from air and fuels, like isooctane and methane, has been studied in a cell as well as in a four-stroke spark ignition engine. Especially, spectral interference from commonly used fluorescent additives (3-pentanone and acetone) with the Raman signals has been investigated. Moreover, possibilities for suppressing the background originating from e.g. lubricants or fluorescent tracers, by taking advantage of the polarisation characteristics of Raman scattering has been studied briefly. Finally, demonstration measurements of the calibration technique have been performed and the precision and accuracy of the method have been analysed.

The LIF/Raman-calibration is showing promising results and the next step will be to apply the technique in an industrial application. [Richter et al., 1998b]

**Optical characterization of Dimethyl Ether (DME) for laser-based combustion diagnostics**

Ö. Andersson, H. Neij, J. Bood, and B. Axelsson

As a result of tightening emission standards, especially for nitric oxide (NO) and particles, compression ignition (CI) engines attract much attention. There is an inherent dilemma in the simultaneous reduction of both these emissions, since NO production is favored by long residence times and high temperatures, conditions under which soot is oxidized in the cylinder. The investigation of CI engine combustion by means of optical methods are limited due to the occurrence of particles and the fact that diesel absorbs heavily throughout the UV region. Lately, there have been reports of the use of dimethyl ether (DME) as a substitute fuel for diesel. Operating CI engines with DME is claimed to reduce NO emissions by 75% and virtually eliminate soot, meeting the California Ultra Low Emissions Vehicle (ULEV) regulation for medium duty vehicles. The thermal efficiency is equivalent to diesel operation. This obviously makes DME interesting both as an alternative fuel and as an aid for making optical investigations of combustion processes in CI engines.

DME is a well-known substance and is for example used as propellant for spray cans, since it is non-toxic and environmentally benign. Recently it has been found that DME can be produced from natural gas in large quantities at low cost. This implies that DME may become important as an alternative fuel in the future. Reports on its optical characteristics are however sparse in the literature. DME was therefore investigated by flame-emission, optical absorption, laser-induced fluorescence (LIF), Raman spectroscopy, and rotational CARS.

Although the peak of the first absorption band of DME resides around 184 nm, significant absorption can also be expected in the long-wavelength tail above 190 nm. It is mainly in
this region that tunable laser radiation is available. At 192 nm the room-temperature absorption cross-section is 7.2 ± 1.5 Mbarn. Even in a lean fuel-air mixture, typical for CI engines, this high absorption cross section will result in complete depletion of a 193 nm laser beam over a fraction of the cylinder diameter. It is thus important to use a laser wavelength in a spectral region where the mixture is optically thin. The edge of the absorption band is displaced towards longer wavelengths with increasing temperature at a rate of about 0.02 nm/K, within the investigated temperature interval.

A free flow of DME in air shows several fluorescence bands in the 350-550 nm region, upon illumination at 193 nm. The LIF signal is of the same order of magnitude as the spontaneous Raman scattering from DME and decreases with temperature. Excitation at 230 nm yields a faint fluorescence in the 280-320 nm region. Excitation at 248 nm yields no detectable fluorescence. Detecting DME by LIF is impractical using 193 nm excitation due to the strong absorption and relatively low signal intensity. Excitation at longer wavelengths yields even lower signals. When measuring other combustion species than DME, this might be a favorable property.

A relative spontaneous Raman cross section of DME is determined to be 2.2 ± 0.2 times the value for methane at 248 nm. The vibrational Raman shift of DME is 2900 cm⁻¹, and the Raman line is thus well separated from the laser wavelength. These properties imply that spontaneous Raman scattering may be an especially useful diagnostic technique in environments containing DME.

The rotational CARS signal from DME is about ten times weaker than the signal from molecular nitrogen. DME’s non-resonant susceptibility is about nine times greater than

Figure 3.24: Absorption spectra for DME. The absorption is displaced towards longer wavelengths with increasing temperature. A methane spectrum is shown for comparison.
that of molecular nitrogen. Temperature measurements by rotational CARS can be performed in DME/air mixtures containing up to 40% DME (a stoichiometric DME-air mixture consists of about 7% DME). Simultaneous relative concentration measurements are not feasible at the moment, for several reasons: DME is a difficult molecule to model— the polarizability tensor is to our knowledge not available. Furthermore, as the rotational lines of DME are not possible to resolve with the spectrometer used (f = 1.0 m, 600 lines/mm) the sensitivity might be quite poor. However, since the vibrational Raman cross section is shown to be comparatively strong, one may expect a strong vibrational CARS signal. A possible approach for making simultaneous temperature and concentration measurements for DME fractions up to 40% could therefore be to combine rotational and vibrational CARS using a double-folded BOXCARS configuration. In such an experiment the shape of the rotational CARS signal is used for temperature measurements and the vibrational CARS signal is used for evaluation of the DME fraction by measuring the ratio between the DME and nitrogen signals. However this approach needs accurate calibration measurements and the dependence of both the rotational and vibrational CARS signal intensities on temperature has to be known.

Preparations for laser diagnostic investigations in a DME fueled direct-injection CI engine are presently underway at our laboratories. [Andersson et al., 1998]
4. SPARK IGNITION AND ION CURRENT SENSORS

4.1 Ion Sensor

A. Franke, R. Reinmann, and A. Saitzkoff

Measurements of what is going on inside an internal combustion engine is a challenging task. The use of advanced laser techniques has been successful in many instances, but is not very practical for monitoring the inside of an engine during regular use in motorized vehicles. Utilizing something already present in the combustion chamber would be of great advantage, not only to minimize the total cost but also the number of sensing probes which could disturb the combustion process.

By applying a small voltage across the spark gap after the spark discharge, a small current due to ions in the combustion gas can be generated. A typical current trace is shown in Figure 4.1. This current is obviously related to the concentration and mobility of charged particles in the gas, which can be generated by two main mechanisms. During the early phase of flame development, the reaction zone passes the electrodes, thus supplying a path for the current with the help of charged particle generated mostly by chemi-ionization (peak c in Figure 4.1). Later on, as the pressure and the temperature rise, thermal ionization in the burned gas provides the free charges required for a current between the electrodes (peak d in Figure 4.1).

![Figure 4.1](image_url)

**Figure 4.1:** A typical ion current and the corresponding pressure trace. The first peak (c) after coil on/off (a/b) occurs due to chemi-ionization in the vicinity of the spark gap and carries information about ion concentrations in the reaction zone. The second, much larger peak coincides with the maximum pressure and is due to thermally created ions.
Currently, ionization sensors are used for knock and misfire detection. Both can be achieved by electronically analyzing the current signals magnitude and frequency spectrum [Auzins, 1995]. In [Reinmann, 1997], the capability of the ion current to predict the air-fuel equivalence ratio, $\phi$, which is usually determined with the help of an oxygen sensor placed in the exhaust gases, has been investigated both experimentally and theoretically. That work is focussed on the first ion current peak which is a result of chemi-ionization in the flame front. A steady state analysis of that process has been done. A major result of that modelling was that the concentration of the hydronium ions should scale as

$$[H_3O^+]_{\text{max}} \propto \sqrt{\phi} \quad (4.1)$$

The concentration of electrons which carry most of the ion current has been shown to be directly related to the concentration of the hydronium ions, thus enabling the calculation of the first peak if an electric field can be assumed. Those calculated first peaks are shown together with data obtained from engine measurements in Figure 4.2.

The second ion current peak, related to thermal ionization in the burned gas, has been explored in a further project [Saitzkoff, 1997]. The modelling of that process yielded e.g. information about the importance of electrons, positive and negative ions, and enabled the calculation of the pressure trace from the ion current and vice versa. A result of that mapping is shown in Figure 4.3. Closely related is the calculation of the heat release from the second ion current peak. A method to perform this has been suggested in [Reinmann, 1998], which was focused on fuel additives and their influence on the ion current. Figure 4.4 shows results of the attempt to correlate the heat release to the ion current. It is obvious that the ion current can be used to derive heat release values which are close to 100%, but is mostly useless for the determination of heat release values early in the combustion.

**Figure 4.2:** The ion currents, calculated from the ion concentration achieved from the chemical kinetic model, are indicated by symbols, and the measured ion currents are represented by the lines according to the legend.

**Figure 4.3:** A correlation between predicted and measured peak pressure for a few different driving conditions.
In order to take a closer look at the capabilities of the ion current to predict the local air-fuel ratio, the following experiment was designed and conducted [Neij,1998]. The usual way to verify a new technique is to compare the results obtained by it with the ones obtained by a well-known technique. In our case, the object of interest was the local air-to-fuel equivalence ratio. Information on the global value of $\phi$ can be acquired via an oxygen sensor as mentioned above. Spatially and cycle resolved $\phi$ maps are obtainable utilizing two-dimensional laser induced fluorescence. The strategy applied was as follows: ion current and $\phi$ data were acquired as well as LIF maps, showing the fuel concentration in relative values. The maps were calibrated using the global $\phi$ value from the oxygen sensor. Then the equivalence ratio around the spark plug could be picked from the $\phi$ map and correlated to certain features in the ion current. The setup needed to conduct those measurements simultaneously is shown in Figure 4.5. Two essential parameters were varied during the experiments: the equivalence ratio, $\phi$, and the mixing time. For the variations of $\phi$, the mixing time was chosen to be long enough to guarantee a homogeneous mixture. The mixing time was varied while keeping $\phi$ at a constant value of 1.

The equivalence ratio could successfully be correlated with several features in the ion current, for example the current peak values and the integral of the two peaks as well as the delay between spark and the occurrence of the first and second peak in the ion current. This holds even for individual cycles. However, the current delay exhibits a highly nonlinear dependence on $\phi$ which makes an unambiguous $\phi$ mapping from that feature impossible. A combination of current delay and peak or integral values might however yield sufficiently reliable information about the local air-to-fuel equivalence ratio.

As for shorter mixing times, no clear dependence of the current delay could be revealed. This might be due to the two counteracting phenomena, i.e. flame speed enhancement
due to turbulence versus flame speed decrease due to inhomogeneities. Another problem occurring during the measurements was the signal level of the ion current, in particular the amplitude of the first current peak, which turned out to be much weaker for the spark plug and fuel (methane) used in this case than it usually is in engines. As a consequence of this, the experiment is going to be repeated using both different fuel and spark plug constructions.

### 4.2 Spark Energy Deposition

*A. Franke, R. Reinmann, and A. Saitzkoff*

The amount of thermal energy deposited in a combustible gas mixture by a spark is known to be a crucial point for a successful ignition as well as for the subsequent development of the flame kernel. If ignition fails, a so called misfire occurs which decreases performance and deteriorates drivability. Furthermore, an excess of spark energy can stabilize the flame kernel.

A usual method to determine the spark energy is to measure voltage over and current through the electrodes during the discharge. By integration over the product, a value for the electric energy can be obtained. However, only a part of it actually arrives in the gas. During the different phases of the spark development, the conversion efficiency from electrical to thermal energy of the gas varies significantly. Two ignition systems yielding the same electric energy do not necessarily deposit the same amount of thermal energy in the gas mixture. A more accurate method to determine the real amount of thermal energy is to employ calorimetry. Since the spark energy is in general low (in the order of a few mJ), the sensitivity of a calorimetric method is higher if the heat capacity of the calorimeter is low. That can be achieved by using a sufficiently small chamber for the measurement. The property measured is the pressure rise, from which the thermal energy can be calculated if adiabatic conditions are assumed.
In order to obtain information about the thermal energy deposited in a gas mixture by a spark, five different commercial ignition systems were investigated under varying pressure conditions. Both inductive and capacitive systems were chosen for this experiment, which is described more detailed in [Saitzkoff, 1998]. The acquired data makes it possible to extract not only the true thermal energy, but also gain information about the efficiency of the glow phase and the breakdown energy, which are generally difficult to obtain. A comparison of the measured electric and calorimetric energies can be seen in Figure 4.6.

**Figure 4.6:** The relation between the electric and the deposited (calorimetric) energies for two different ignition systems. Note that even if the system on the right releases a lot more energy the actually deposited energy increases only marginally.
5. SIMULATION OF PULSATING COMBUSTION

S. I. Möller

Pulsating combustion is a combustion process where properties such as velocity, pressure etc vary periodically in time. The oscillations appear spontaneously due to interaction between the flow dynamics and the combustion process. Pulsating combustion has certain advantages compared with traditional combustion techniques, e.g. low emission of pollutants and efficient utilisation of the fuel. For more details regarding pulsating combustion see [Möller, 1997a].

A project about pulsating combustion was initiated in 1989 at Lund Institute of Technology. This project is a collaboration between the divisions of Combustion Physics, Mechanics and Energy Economics and Planning. The project is divided into two main parts, fundamental research and investigation of new applications for pulsating combustion and is carried out with a close collaboration between the theoretical and experimental parts of the project.

Figure 5.1: Measured and simulated pressure cycle for the three flame holder positions (top). Measured relative OH intensity and simulated relative heat release for the same flame holder positions (bottom).
The fundamental part aims at getting increased knowledge of the fundamental interacting physical processes by means of detailed numerical simulations and advanced measurements. In the applied part of the project four separate applications are investigated [Möller, 1997b]

i) Natural gas fired domestic heater
ii) Cofiring larger furnaces with pulse combustors
iii) Pulse combustors of Rijke-type
iv) Pulsating combustion for drying applications

The major contribution from Combustion Physics is within the field of fundamental research on pulsating combustion, particularly on detailed numerical simulations. Numerical simulations can serve as a powerful tool for understanding of the physical processes in a pulse combustor and may also give guidelines on how to design a pulse combustor with certain operating characteristics. The simulation model used here is a general model for time dependent turbulent flow of chemically reacting mixtures. The model is based upon the theory of mixtures within continuum mechanics and utilise the Large Eddy Simulation (LES) concept. The Smagorinsky model is used for modelling of the sub-grid scale stresses and fluxes. The chemical reaction rates are estimated from the kinetically controlled reaction

Figure 5.2: Simulated (top) and measured (bottom) temperature distributions in the pulse burner.
rates and the mixing rates. Velocity field, pressure, temperature and species concentrations are output parameters from the model. A submodel for the formation of nitrous oxides is included in the model. The simulation model is implemented into the CFD-code Phoenics, CHAM Ltd, London, [Möller, 1997a].

For validation of the model simulations of pulse combustors of Helmholtz type corresponding to experimental pulse combustors designed and operated at the division of Energy Economics and Planning have been carried out. These pulse combustors have combustion chambers with walls of quartz glass in order to have optical access. Natural gas is used as fuel. Simulated velocity and heat release fields have been validated against LDV measurements and chemiluminescence from the CH-radical respectively. Results show that vortex structure in the combustion chamber largely influence the combustion process. Validation show that the predicted temporal and spatial distribution of the heat release fields is in good agreement with the emission from the CH radical, [Lindholm and Möller, 1997]. In another study, [Möller and Lindholm, 1998], the ability of the model to predict the operating characteristics for different inlet geometries was investigated. Here the pressure and heat release cycle was investigated as well as tail pipe temperature and O₂ and NOₓ concentrations at the outlet of the pulse combustor. It was found that the timing between the heat release and the pressure was affected by the different inlet geometries (see Figure 5.1). Also the emission of NOₓ was changed. The model was found able to predict the different combustion characteristics both in terms of the timing of the combustion cycle as well as the emission levels.

The temperature field in the combustion chamber has a large influence on the NOₓ formation. Therefore it is very important that combustion chamber temperatures are predicted accurately. In a measurement campaign carried out in collaboration with J. Engström, Combustion Physics, and A. Lindholm, Energy Economics and Planning, the temperature field was measured with two-line atomic Laser Induced Fluorescence, LIF (see section 3.1.2). The data is currently under evaluation. Another example of calculated and measured temperature distribution is shown in figure 5.2.

Combustion Physics has also participated in the applied project (ii), where three pulse combustors of Helmholtz type have been installed in a waste furnace operated by SYSAV, Spillepengen, Malmö. The aim of this experiment is to use the pulsating jets to change the flow field in the large furnace and thereby increase the mixing in order to break up regions with high CO contents.

In collaboration with Division of Mechanics a continuum mechanical model for spray combustion have been formulated and investigated, [Palmqvist et al., 1997]. Also a theoretical investigation of the enhanced heat transfer in pulsating flows have been carried out, [Lundgren et al., 1998a&b].
6. CHEMICAL KINETICS

6.1 Applied kinetic calculations

NO\textsubscript{X} formation

D. Nilsson, T. Lövås, and F. Mauss

During combustion of fuels containing no nitrogen, NO\textsubscript{X} can only be produced in processes involving atmospheric nitrogen. These fuels are well described with the classical NO\textsubscript{X} reaction mechanisms (thermal, N\textsubscript{2}O, NO\textsubscript{2} and prompt mechanisms). It is well known that NO\textsubscript{X} emissions from these fuels increase with increasing temperature and decreasing fuel-air equivalence ratio (in the relevant range). Secondary NO\textsubscript{X} reduction processes like reburning, thermal DeNO\textsubscript{X} and staged combustion are well documented - also in practical use.

If the fuel itself is polluted with nitrogen, large amounts of NO\textsubscript{X} can be emitted. This fact applies to an extensive range of combustibles, e.g. crude oil, biogas and hazardous waste. For successful modelling of this kind of fuels, information on the decomposition of more complex nitrogen compounds is required in addition to detailed knowledge of the basic NO\textsubscript{X} formation mechanisms.

Staged combustion is a method which has been utilised in heat generation applications for some time, e.g. for NO\textsubscript{X} reduction in large-scale stationary coal furnaces and boilers. For nitrogen-free fuels, staged combustion with ammonia doping of the fuel can be used to reduce NO\textsubscript{X} emissions considerably. Often the staging is made up of an initial stage with

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{fig6_1}
\caption{Calculated and measured NO concentrations in the PSR and after the PFR}
\end{figure}

fuel-rich conditions keeping initial NO production low, and a subsequent stage with air injection at a lower temperature to achieve overall fuel-lean operation.

Monomethylamine (CH₃NH₂) can be used as a model compound of fuel-bound nitrogen found in biofuels and waste products. Different kinetic models have been validated using measurements on a scaled-down model of an industrial rich-lean-staged burner fuelled with ethylene (C₂H₄) doped with CH₃NH₂. The burner consists of a toroidal jet-stirred reactor followed by a tubular section with secondary air injection. A successful theoretical model for this device is given by a combination of a perfectly stirred reactor (PSR) and a plug flow reactor (PFR).

NO concentrations simulated with various chemical schemes are compared with measurements in the graphs below, and a good overall agreement is manifested. A clear minimum in NO emissions (after the PFR) for slightly fuel-rich PSR conditions is well reproduced. The successfully validated mechanisms will be further improved using NH₃/NO flame measurements, and subjected to automatic reduction to investigate the potential for application in large-scale simulations of practical devices. [Nilsson et al., 1997, Mauss et al., 1998a]

Modelling of NOₓ and Soot Formation in Turbulent Combustion

A.S. Dederichs, M. Balthasar and F. Mauss

An approach to model the formation and oxidation/reduction of soot and NO in turbulent diffusion flames has been developed. The model is based on the flamelet library approach and was extended to account for radiative heat losses in the flame. Due to the rather slow processes leading to soot and NO a modified flamelet library approach is used. The different source terms for soot and NO formation are calculated and a transport equation for the mean massfractions is solved in the CFD calculation. The source terms are obtained from laminar counterflow-flame calculations using a detailed chemistry model for the gas phase species and the formation and oxidation of soot. Transport equations for the mean mixture fraction and the mixture fraction variance are solved and the chemical source term is closed by presuming a beta-function like distribution of mixture fraction and a log-normal distribution of the scalar dissipation rate. The model was first tested in laminar [Balthasar et al., 1996] and turbulent jet flames [Bai et al., 1998]. By applying a reduction strategy [Balthasar et al., 1997] for the flamelet libraries of the source terms it was made applicable to the simulation of soot formation in a Diesel spray taking different oxidiser temperatures and pressures into account [Karlsson et al., 1998]. Additionally, different formulations of the flamelet equations have been tested and their accuracy has been evaluated by comparing them to turbulent flame experiments.

THE DETAILED SOOT MODEL - The soot model used in the calculations relies on a detailed description of the physical and chemical processes developed by Frenklach and further developed by Mauss [Mauss 1997] (see Figure 6.2). It can be subdivided into the gas phase reactions, the growth of polycyclic aromatic hydrocarbons (PAH) and the processes of particle inception, surface growth, condensation and oxidation. The gas phase
reactions are modelled using a detailed chemical reaction mechanism for the combustion of heptane. It includes 89 species and 855 reactions and, is based on the mechanism of Chevalier et al. modified with recent kinetic data. The different formation and destruction pathways of NO are included in the mechanism. Starting with benzene, repeating cycles of hydrogen abstraction and acetylene addition that can be modelled as a fast polymerisation process describe the growth of PAH. Processes for particle inception, soot growth and oxidation are described by chemical and physical models. Physical models are applied to all coagulation processes, e.g. the coagulation of two PAH forming the first soot particles, the coagulation of PAH and soot particles describing the condensation of PAH on the soot particle surface. A chemical model is applied to the heterogeneous reactions occurring on the soot surface. Most of the soot mass growth is completed by surface growth of the soot particles by reaction with acetylene adding new aromatic rings in a similar manner as the PAH growth (HACA mechanism). The soot is oxidised by reaction with molecular oxygen and hydroxyl radicals. More details on the soot and chemistry modelling are given in [Mauss 1997] and references therein.

FLAMELET CALCULATIONS – The flamelet concept proposed by Peters was applied to uncouple the flow calculations from the combustion processes. Assuming fast chemistry and a thin reaction zone, equations for energy and species conservation are formulated as a function of the mixture fraction and the scalar dissipation rate that accounts for non-equilibrium effects. The flamelet equations are obtained by a co-ordinate transformation from the physical into the mixture fraction - scalar dissipation rate space of the governing equations for a counterflow-flame. These equations can be simplified by determining the leading order terms with the help of an asymptotic analysis. To evaluate the accuracy of different assumptions leading to the classical flamelet equations the source terms of soot and NO formation and oxidation calculated for the fully transformed set of flamelet equations were compared to the solutions of the simplified equations [Dederichs et al., 1998]. Best results were obtained assuming unity Lewis numbers in the outer layer of the flamelet and non-unity Lewis numbers in the thin reaction layer. Solutions obtained with different flamelet equations for the source term of surface growth are given in Figure 6.3.

![Figure 6.2: Illustration of the different processes involved in the formation and oxidation of soot.](image-url)
The flamelet library approach for the sources of soot formation and oxidation has been tested in a laminar C\textsubscript{2}H\textsubscript{2}/Air jet flame first [Balthasar et al., 1996]. The accuracy of the reduced flamelet library approach was evaluated in a CH\textsubscript{4}/Air laminar jet [Balthasar et al., 1997]. The approach was consecutively applied to a turbulent C\textsubscript{2}H\textsubscript{4}/air jet flame [Bai et al., 1998] including the radiative effects in the flamelet calculations. Turbulence was modelled with the k-\(\varepsilon\) model while combustion was modelled via the flamelet approach. The soot volume fraction is calculated to be in good agreement with the measurements as shown in Figure 6.4.

Soot formation and oxidation was simulated in a spray burning under Diesel engine conditions [Karlsson et al., 1998]. Combustion was modelled by an eddy break-up model. The

**Figure 6.4:** The calculated soot mass as function of CAD for different injection timings (left), calculated soot volume fraction at different heights above the burner compared to measurements in a turbulent C\textsubscript{2}H\textsubscript{4}/Air jet flame (right).
soot model could be implemented in the CFD code without major changes being necessary to the main codes. The soot model part of the code consists of a separate subroutine in which the detailed flamelet library is summarised in the form of simple analytical functions. The modifications to the code resulted in only a minor increase, approximately 20%, of the overall CPU time required for a run. Three engine operating conditions, differing mainly by the injection timing, were used in order to study the performance of the soot model. These conditions all correspond to part load conditions for a Volvo D12A truck engine. The experimental data used in this work were obtained during measurements using a one-cylinder research version of the Volvo D12A engine. It is interesting to note that the results obtained by calculations are in qualitative agreement with the soot distributions measured by Dec and co-workers for similar engine conditions. This means that maximum soot volume fraction is found in a broad area in the tip of the spray. At later times, however, the maximum soot volume fraction occurs at positions where the spray was evaporated, see Figure 6.5. Here the gas temperatures are lower than in the tip and the oxidation of soot is slower.

Results shown in Figure 6.4 illustrate the time evolution of the total soot mass for the three operating conditions. The calculations were terminated at a crank angle of 420 degrees. However, qualitatively by extrapolation, it can be observed that the calculated amounts of soot are in the correct range. Although the correct trends are predicted, the calculated final soot masses seem to converge towards a single final soot mass. This is in contrast to the measured results showing significant differences in exhaust soot masses. Since the presence of walls and other 3-d effects are not taken into account in the calculations full geometry calculations have to be performed to get a better insight on the predictive capacities of the model.

Figure 6.5.: Distribution of the soot mass in the spray at CAD 386
The Influence of Microscale Mixing on Soot and NO\textsubscript{x} Formation

M. Balthasar, M. Kraft\textsuperscript{*} and F. Mauss
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The emission of nitrogen oxides and soot from combustion sources into the atmosphere causes severe detrimental effects on the environment. To reduce NO\textsubscript{x} and soot emissions practical models for industrial combustion devices such as gas turbines have to be developed. In most cases industrial burners work under turbulent conditions. While the detailed model to trace the NO\textsubscript{x} and soot emissions presented above [Balthasar et al., 1997] is valid in the flamelet regime we focus in this work on the distributed combustion regime.

On the basis of the stochastic reactor models Partially Stirred Plug Flow Reactor (PaSPFR) and Partially Stirred Reactor (PaSR) a model is developed that accounts for soot and NO\textsubscript{x} formation in the distributed combustion regime [Mauss et al., 1998b, Kraft et al., 1999]. This is done by means of the joint composition and joint moments of soot particle size probability density function. The gas phase reactions are modelled by a detailed chemical mechanism containing 104 chemical species. This model is used to calculate the second stage of a bench scale combustion unit. Two different cases are studied. In the first case the model predictions are compared to measurements performed in the reactor. The sensitivity of number density and soot volume fraction on the variation of the mixing intensity is studied in the second part.

A complex model was used to describe the chemical and physical processes occurring during combustion [Mauss, 1997]. The processes can be divided into three parts: the gas phase reactions of species up to small PAHs (polycyclic aromatic hydrocarbons), the growth of PAHs and the formation and oxidation of soot. Equations for the first four moments of the soot particle size distribution function are solved:

---

**Figure 6.6: A schematic view of the combustor**
\[
\frac{\partial' M_y}{\partial t} + \frac{\partial(U_j' M_y)}{\partial X_j} = \frac{\partial}{\partial X_j} \left( \rho D' \left( \frac{\partial M_{y+1/2}}{\partial \psi_i} \rho \right) \right) + \rho S_{y\psi_i,\rho} \left( r = 1, \ldots, N_r \right) \tag{6.1}
\]

The first moment represents the soot particle number density \( N \) whereas the second moment is proportional to the soot volume fraction \( f_v \).

The partially stirred plug flow reactor model (PaSPFR) is the stochastic equivalent of the plug flow reactor model (PFR) based on the PDF approach. The time evolution of the scalar statistics is modelled in a volume element flowing with a mean velocity along the reactor axis for a given set of initial conditions. The scalar \( \phi \) is assumed to be a random variable: 
\[
\phi(\psi; t) = (Y_1, \ldots, Y_{104}, M_0, M_1, M_2, M_3, \ldots, t)
\]

\[
\frac{\partial \phi(\psi; t)}{\partial t} + \frac{\partial}{\partial \psi_k} \left( S_k (\psi) \phi(\psi; t) \right) = \frac{\partial}{\partial \psi_k} \left( \frac{1}{2} C_0 \left( \psi_k - \bar{\psi}_k \phi(\psi; t) \right) \right) \tag{6.2}
\]

Mixing in the combustor is modelled via the Binomial Longevin mixing model in case 1 and via the IEM model in case 2. Statistically homogeneous and isotropic turbulence is assumed.

The combustor studied consists of a toroidal jet-stirred combustor (TJSC) followed by a turbulent plug flow reactor as displayed in Figure 6.6. The TJSC is used to generate a well-controlled homogeneous mixture of hot gases entering the turbulent plug flow reactor. The combustion products pass through a flow straightener and then through a turbulence grid to generate a turbulent flow. The turbulence time scale has been calculated as \( \tau = l_i / u' \).

Two different cases were studied. For both cases the TJSC was operated with a mixture of \( \text{C}_2\text{H}_4 \), air and \( \text{N}_2 \) (\( \phi = 0.66 \)) with an exhaust temperature of \( T = 1390\text{K} \). In the first case (case 1) \( \text{C}_2\text{H}_4 \) diluted with nitrogen at \( T = 1200\text{K} \) was injected into the hot exhaust stream. The molar flow rate ratio of injection to exhaust stream was 5\% (\( \dot{n}_i / \dot{n}_e = 0.05 \)). Measurements were performed for the molefractions of \( \text{C}_2\text{H}_4 \) and \( \text{C}_2\text{H}_2 \) and compared with the simulations. This case was used to check the predictive capacity of the model. Since no soot was formed under these conditions a second case (case 2) was simulated choosing conditions typical for the formation of soot. Pure \( \text{C}_2\text{H}_2 \) at \( T = 1000\text{K} \) was added to the hot
combustion products. The molar flow rate ratio was increased to $n_i / n_e = 0.12$. The effect of different mixing rates on the number density $N$ and the soot volume fraction $f_v$ was studied. Equation 2 has been solved using a particle method as described in [Kraft *et al.*, 1999]. The statistical moments of the scalars can be estimated from the particle ensemble. The mean molefractions of $C_2H_2$ and $C_2H_4$ for case 1 compared to the measurements are shown in Figure 6.7. The imperfect mixing leads to a much slower decrease of the $C_2H_4$ molefraction and a higher concentration of $C_2H_2$, a key species for soot formation, as expected for perfect mixing.

Mean number density and soot volume fraction for different mixing intensities are shown in Figure 6.8 ($\tau=0$ corresponds to the perfect mixed case). Very low soot volume fractions and number densities are found for the perfect mixed case. For higher values of $\tau$ number densities increase and reach a local maximum. This maximum is shifted to higher residence times for increasing $\tau$. The number density reaches the same value at the outlet for all mixing intensities due to the coagulation of the soot particles. The soot volume fraction increases by two orders of magnitude with decreasing mixing intensity.

Decreasing the mixing intensity small amounts of hot exhaust coming from the TJSR are mixed with the injected fuel resulting in ideal conditions for soot formation. Soot is not oxidised due to the lack of oxygen in the later stage of the combustor. It can be concluded that the number density at the outlet is not affected by increasing $\tau$ whereas the soot volume fraction is increased by two orders of magnitude. Thus the same amount of soot particles is emitted for all cases with the soot particles increasing in size for increasing $\tau$. The concentration of NO increases dramatically when decreasing the mixing intensity due to a faster rise in temperature. The result of this sensitivity study stresses the importance of microscale mixing for the prediction of soot formation in turbulent flames.

*Figure 6.8: Mean number density and mean soot volume fraction.*
Numerical Simulation of a Homogeneous Charge Compression Ignition Engine

Per Amnéus and Fabian Mauss

The homogeneous charge compression ignition engine (HCCI-engine) is often considered as the third alternative to spark ignition engines (SI-engines) and compression ignition engines (diesel engines). Here, a homogeneous charge is used as in a SI-engine, but the charge is compressed to auto-ignition as in a diesel. The ignition process is described in Figure 6.10. The main difference compared with the SI-engine is the lack of flame propagation and hence the independence from turbulence. Unlike the diesel engine, the combustion is premixed, and does not have the problems with soot formation associated with diesel engines.

The ignition process of an HCCI-engine was simulated, using a mathematical model, basically consisting of zero-dimensional time-dependent differential equations. The energy conservation and mass conservation balance for each chemical species is solved with Newton’s method and the time is resolved with higher order backward differential functions. The main characteristics of the program have been discussed in several papers [Hajireza et al., 1997, Amnéus et al., 1998]. The detailed kinetic mechanism used for the

Figure 6.19: Schematic view of the autoignition process in a HCCI-engine. (Upper left:) Intake of a premixed air-fuel mixture. (Upper right:) Compression of the premixed charge. (Lower left:) Compression to autoignition at hot spots in the combustion chamber. (Lower right:) Immediately thereafter; the entire bulk volume is enflamed.
simulation of the ignition process contains 64 species and 832 reactions. This mechanism was adapted to fit the lean fuel/air mixtures and high pressures that are present in HCCI auto-ignition processes.

Simulations were made, obtaining the initial pressure and the temperature from the experiments at a specified crank angle position. The simulations show that an acceptable agreement could be obtained with this very simple mathematical model, independent if the engine was running naturally aspirated or supercharged [Cristensen et al., 1998], as shown in Figure 6.10. The result of the calculations also suggest that the impact on the ignition timing from variations in temperature is very high, whereas the impact from variations in pressure and fuel/air ratio is less dramatic [Amnéus et al., 1998]. The calculations also suggest that, when using natural gas as fuel, the composition of the fuel, with respect to higher hydrocarbons such as ethane, propane and butane, have a non-negligible effect on the ignition. As can be seen in figure 6.11, this effect is very evident when the engine is running under conditions close to ignition limit.

6.2 Fundamental kinetic investigations

Complex Fuels and their Effects on Engine Knock

*Per Amnéus, Jesper Rönnbäck, and Fabian Mauss*

Engine knock is a highly undesired phenomenon in spark ignition engines, as it might cause damages to the engine. It is started by auto-ignition in the end-gas, caused by the pressure rise from the piston movement and from the flame propagation. The further development from auto-ignition to engine knock requires that the speed of combustion is faster than the flame velocity.
Calculation of the auto-ignition process that incites engine knock has been performed, using a two-zone model [Hajireza et al., 1997, Bood et al., 1997] and a 3-zone models [Hajireza et al., 1998]. In the two-zone model, the approach is to divide the combustion chamber in a burned gas zone and an end-gas zone, where the flame is calculated as a disturbance between the burned gas zone and the end-gas zone. In the 3-zone model, a zone for the unburned gas in a thermal boundary layer is added.

The mentioned calculations uses a semi-simplified kinetic mechanism scheme for primary reference fuels (PRF-fuels), where a skeleton mechanism for n-heptane and iso-octane is added to a detailed mechanism for C$_7$- C$_4$ hydrocarbons. These calculations show an acceptable agreement, although improvements are desirable, improvements that might be obtained by a more sophisticated mathematical model, and by a more detailed kinetic mechanism.

In real life, engines are not fuelled n-heptane/iso-octane mixtures, but by complex fuels, consisting of a few hundred different fuel components. Some of these components are aliphatic, others aromatic. These complex fuels have in some extent different knock characteristics than PRF-fuels of the same octane rating.

A current task is to develop new kinetic reaction mechanisms, and to adjust mechanisms to fit different demands, such as different temperature or pressure regions. In some cases, a less detailed mechanism is desired, to minimize the computing time. Sensitivity analysis and reaction flow analysis are two commonly used tools that enables adaptations and reductions of kinetic mechanisms.

**Automatic Reduction of Detailed Chemistry**

*T. Lövås, D. Nilsson, and F. Mauss*

There are several ways of reducing the chemical complexity of a combustion system. The reaction mechanism reduction techniques rely on the fact that many of the chemical combustion processes reach equilibrium on a time scale that is much smaller than the time scales that are relevant for a combustion process.

For some set of conditions, steady-state and partial equilibrium equations that describe most of a detailed reaction mechanism without substantial loss of accuracy can be derived by manual calculation. Quite some calculation effort is required for obtaining the minimum dimension and the rate constants for a conventional reduced mechanism. The reduction procedure has to be repeated for different geometries and physical and chemical conditions, and often the validity of the simplifying assumptions is not evident until the reduction is finished and actual simulations have been made. Therefore it is an attractive idea to automate the reduction process. This can be done by applying systems analysis methods to the linearized system of differential equations describing the time dependence of species concentrations, temperature and pressure. A measure of species lifetimes is extracted from
the jacobian matrix (i.e. the derivative matrix) of the source terms. This species time scale is the life time of the species following a sudden change in its own concentration.

The dynamic behaviour of the species is however made up of contributions from several reactions, and a single time scale which is independent of the concentrations of other species cannot be determined. The species are coupled to each other via their reactions. According to linear systems theory, independent decoupled time scales are instead given by the eigenvalues of the jacobian. Intuitively, one can imagine that we lump together a combination of species that have similar life times and assign a single decoupled time scale to them, thus saving a number of dimensions. A large negative eigenvalue (which is a rate measure) indicates a process with a short time scale, i.e. a quickly equilibrated process. The species combinations with the shortest timescales can now be excluded from the differential equation system one by one, since they are not coupled to each other anymore. We can thus use general mathematical methods to discriminate between processes, describing the fast processes with simple algebraic equations, analogous to the steady-state and equilibrium expressions of the manually reduced mechanisms. This principle is the theoretical foundation of the methods termed the Computational Singular Perturbation (CSP) method and Intrinsic Low Dimensional Manifold (ILDM) method. The ILDM method involves the computation and tabulation of fast species concentrations as functions of a small number of slow ones.

**Figure 6.2:** Combustion of different stoichiometric CO-H2-air mixtures in a plug flow reactor. (left:) Chemical dimension of one of the mixtures estimated by number of ILDM conditions that are not fulfilled (right axis), and temperature evolution (left axis) as functions of residence time. Black graph represents C2 chemistry, and the grey graphs are results with C4 and NOx chemistry added. (right:) H2O concentration versus CO2 for all residence times and all sets of initial conditions. White points have dimension numbers <3 and can be seen to coincide with a one-dimensional manifold.
To illustrate the way to ILDM computation, a plug flow reactor fuelled with different mixtures of CO, H\textsubscript{2} and their products is used as a simple example. In the left plate of fig 6.2, the number of significant processes, i.e. the effective chemical dimension is plotted as a function of residence time. The chemical dimension is estimated by the number of vectors of the Schur decomposition of the jacobian that are non-orthogonal to the source terms (It can be shown that this orthogonality condition is applicable to eigenvectors or Schur vectors, and signifies the fast relaxation of a process). Close to ignition there are hardly any negligible species or reactions, and the number of non-relaxed processes approaches the number of species in the detailed mechanism. Before ignition and for longer residence times, closer to equilibrium, the dimension number is low as expected, and should coincide with an ILDM.

To check this, the H\textsubscript{2}O concentrations for all residence times and different initial conditions is plotted against the CO\textsubscript{2} concentration in the right plate of Figure 6.2. In spite of the varying initial conditions, the H\textsubscript{2}O concentrations follow essentially the same path to the same equilibrium point, and should be adequadly described by a one-variable algebraic equation for longer times. The white points are the ones with low dimension numbers (<3) from the left graph. This 1-dimensional part of the plot is the intrinsic Low-Dimensional Manifold, which can obviously be pinpointed by our calculations.
6. REFERENCES


Kraft, M., Balthasar, M., and Mauss, F., “Soot and NOx Formation in a Stationary Turbulent Combustor”, to be published in the proceedings of the International Workshop on


