Oxy-fuel combustion analysis in an industry relevant furnace via vibrational CARS thermometry

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Abstract

The coherent anti-Stokes Raman spectroscopy (CARS) technique is a very well suited laser based tool for a non-intrusive investigation of turbulent high temperature combustion processes. In this work we analysed with the help of O_2 based vibrational CARS system an industrial 400kW oxy-fuel burner which is integrated in an industrial relevant test furnace. The burner is feed with pure oxygen and natural gas at an equivalence ratio of ϕ=0.9. At one downstream position temporal and spatial resolved temperatures were measured along a 600 mm line. Additional air sucked in from the environment seems to influence the gas phase temperature significantly.

Introduction

Combustion processes on an industrial scale in which oxygen instead of air is used as an oxidizer are getting more and more into focus. These so-called oxy-fuel combustion processes are of great interest. They are further developed for instance for the steel and the glass industry [1,2]. In the glass industry of the United States already more than 25 % of all glass-melting furnaces are heated by such oxy-fuel combustion processes [3].
The expression "oxy-fuel combustion" is used in two different ways. In the area of power plant technology the aim is to prevent the release of large quantities of CO_2 into the atmosphere. One possibility is, to combust the fuel in an oxygen-carbon dioxide atmosphere [4,5]. By introducing an exhaust gas recirculation the exhaust gas, which is mostly consisting of carbon dioxide and water vapour is fed back into the combustion chamber to keep the temperature in the combustion chamber on a low level and to rise the amount of CO_2. Thereby a significant increase of the carbon dioxide concentration can be reached which makes its removal from the exhaust gas more effective. This removed CO_2 can be stored e. g. in depleted oil and gas fields which is the basic idea of carbon capture and storage (CCS). In the area of power plant technology the oxy-fuel combustion is one potential means of mitigating the contribution of fossil fuel emissions to global warming.

In thermal process engineering however the oxy-fuel combustion process is used to reach high combustion temperatures even without preheating. In this case the combustion of fuel with pure oxygen as an oxidizer is referred as oxy-fuel combustion. The application of such oxy-fuel burners shows some clear benefits compared to air breathing burners. The amount of NO_x emissions of a pure oxygen combustion processes is significantly lower, which is important in order to fulfil emission regulations. Additionally the combustion with pure oxygen leads to high temperatures without any costs for air preheating. As an example, the calculated adiabatic flame temperature for a stoichiometric methane-air flame (p=1 bar, $T_{surrounding}$=298 K) is approximately 2250 K compared to roughly 3050 K for the corresponding methane-oxygen flame [6]. Although this type of oxy-fuel combustion is increasingly applied in thermal process engineering there is still a need for optimization. For such an optimization the species concentration information and the gas phase temperature is of utmost importance. These data can be used for a validation of numerical models and they are additionally necessary for an improved understanding of the basic chemical reactions and the pollutant formation.

Up to now there are only a few experimental investigations of such industrial relevant oxy-fuel processes available [7-9]. Nevertheless there is still a need for precise experimental data achieved with high spatial and temporal resolution. These data could then also be used for a validation of numerical models. Such precise measurements in industrial relevant oxy-fuel processes like, e.g. in glass melting furnaces are challenging due to the high combustion temperature in combination with a high turbulence. Here laser-based non-invasive measurement methods are in principle well suited to deliver the necessary information (see e.g. [10, 11]). Nevertheless the presence of dust particles, high temperatures near the furnace due to radiation and the availability of only small optical accesses within a large furnace are still limiting the application of most laser based measurement techniques.

Coherent anti-Stokes Raman scattering technique (CARS) offers the possibility for precise temperature measurements in flames (see e. g. [12-14]). It has been developed and used now for some decades and is currently one of the best established non-intrusive thermometry tools in the field of combustion analysis.

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Several studies of different types of flames have proven its high capability [15-18]. Today, the CARS technique using the N_2 molecule as a temperature indicator is routinely used even in harsh combustion environments such as IC engines, high-pressure burners and gas turbines [19-24]. In order to probe temperatures in a technical relevant combustion chamber, optical access is always restricted by the need for very small windows. Since CARS is a coherent technique, it has the advantage that only two, small, line-of sight windows are required to focus the laser beams into the probe volume and to collect the generated signal.

Therefore in this work we present the application of a vibrational-CARS system for gas phase temperature measurements in large scale furnace equipped with a typical industrial oxy-fuel burner. Since in the oxy-fuel process no nitrogen is present the O_2 molecule was used as a temperature indicator.

Experimental

CARS setup

Generally, in CARS the Raman transition of interest is excited resonantly by the frequency difference of two laser beams irradiating the sample, one narrowband e.g. a frequency doubled Nd:YAG (ω_1) and the other (ω_2), a broadband source, usually a dye laser or an optical parametric oscillator (OPO). A third laser beam (ω_3), normally from the same laser source as the first pump beam, is scattered from the excited Raman transitions and the signal is emitted in a coherent, laser-like beam at the anti-Stokes frequency of the probed species. The species dependent CARS signal is generated at frequency $\omega_{CARS}=\omega_1-\omega_2+\omega_3$. The frequency difference between the pump beam ω_1 and the Stokes beam ω_2 has to match a Raman resonance of the molecule of interest which is used for the temperature determination. In air breathing flames N_2 is usually used as a temperature indicator in the CARS process since it is inert. In an oxy-fuel process however there has to be used either O_2, CO or CO_2 as a temperature indicator. We used oxygen for the temperature determination since a lean oxy-fuel flame is investigated. Additionally the vibrational CARS process for diatomic molecules is still easier to model and therefore such CARS signals are simpler to evaluate [25]. A detailed description of the CARS process can be found, e.g. in [10, 26].

A schematic drawing of the experimental vibrational CARS setup is shown in Fig. 1. The laser system is based on a frequency doubled, injection-seeded Nd:YAG laser. The output power was 800 mJ at 532 nm. The pulse duration is 8 ns and the repetition rate is 10 Hz. This Nd:YAG laser provides in the CARS process the first and second pump laser beam (ω_1, ω_3). Part of the 532 nm Nd:YAG laser output is used to pump a commercial broadband dye laser at a central wavelength of 580 nm. The dye laser was equipped with

a mixture of Rhodamine 6G and Rhodamine B in ethanol. The dye laser acts in the CARS process as the Stokes laser (ω_2) and these two dyes were mixed to match the vibrational Raman resonance of oxygen. The laser beams were overlapped in a USED CARS geometry [27]. Here the dye laser beam is centred inside the donut shaped Nd:YAG laser beam. The donut shaped Nd:YAG laser beam is generated by a high reflecting mirror (HM) with a central hole (see Fig 1). In this configuration both laser beams will face the same density changes inside the large furnace and the influence of beam steering is minimized. The laser beams are focused into the probe volume and collimated afterwards by lenses with a focal length of 2500 mm. The resulting measurement volume was determined to be approximately 3.5 mm in length and 150 μm in diameter. For the dye laser a power of 10 mJ and for the Nd:YAG laser 25 mJ was measured in the probe volume. The O_2 CARS signal occurring at 491 nm was separated from the laser beams by dichroic mirrors (DC) and focussed on the entrance slit of a spectrometer. The detection system consists of a spectrometer with a focal length of 550 mm and a grating consisting of 2400 lines/mm. The CARS signal was detected by a charge coupled device (CCD) camera. At each measurement position 1000 single shot spectra were recorded and evaluated using an in house developed CARS code [28]. To take line narrowing into account, the theoretical vibrational CARS spectra were calculated by use of the modified exponential gap law [29]. The parameters for O_2 were taken from Huber at al. and Rouille at al. [30-31].

Burner setup

The oxy-fuel burner was mounted inside a large test furnace which is shown schematically in Fig. 2. The furnace has a width of 1000 mm and a length of 5000 mm. Temperature measurements were performed at different positions along a horizontal line as marked in Fig. 2 1500 mm downstream the burner exit. Due to the axial symmetry the probe volume positions have to cover only half of the combustion chamber. For these CARS measurements two opposite holes in the wall of the combustion chamber were used for the incoming laser beams and for the signal detection. The diameter of the hole for the incoming laser beam was 60 mm. The hole on the opposite site was concentric with an inner diameter of 250 mm facing the combustion chamber. The outer diameter was 60 mm. The measurements were performed without optical windows. For these first measurements a downstream position of 1500 mm was chosen in order to investigate a region where chemical reactions take place.

The industrial diffusion type oxy-fuel burner had a maximum available thermal power of 400 kW. Two independent gas supplies, one for natural gas and one for oxygen were connected to this diffusion burner.
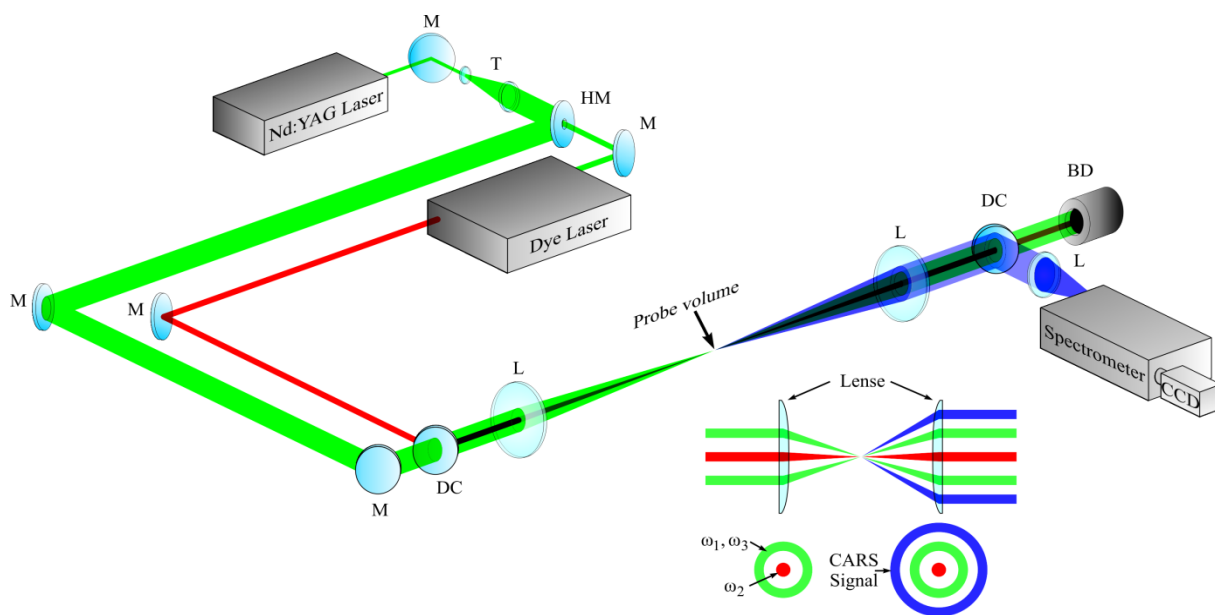
Figure 1:   Experimental CARS setup: M, mirror; T, telescope; HM, hole mirror; DC, dichroic mirror; L, lense; BD, beam dump; CCD, charge coupled devise camera.

Mass flow controllers were used to adjust the flow rates for both gases. A flow rate of 40Nm³/h was set for natural gas and an equivalence ratio of $\phi=0.9$ was used in the experiments shown in this work.

Results and Discussion

The temperature results achieved along the line 1500 mm downstream of the burner are shown in Fig. 3. Measurements were taken each 40 mm. The mean temperature and the standard deviation for 1000 single shot measurements are shown in Fig. 3a. It should be noted that nearly at all measurement positions a large number of spectra could be evaluated. In maximum only less than 21 % of the CARS spectra have to be sorted out probably due to beam steering effects or dust particles. The points at x=-50 mm and x=-30 mm were taken in the concentric hole within the chamber wall and a mean temperature of about 1000 K was evaluated. The highest mean temperature of 1787 K was measured at x=270 mm. The temperature is decreasing towards the centre. At x=510 mm a mean temperature of 1293 K was achieved. Obviously in this part of the furnace the flame is slightly shifted to one side. This is also confirmed by the standard deviation. As an example in Fig. 3 b-d scatter plots of the single shot temperatures are shown. The corresponding probably density distributions are displayed in Fig. 3 e-g. Due to the high turbulence of this oxy-fuel combustion process a large
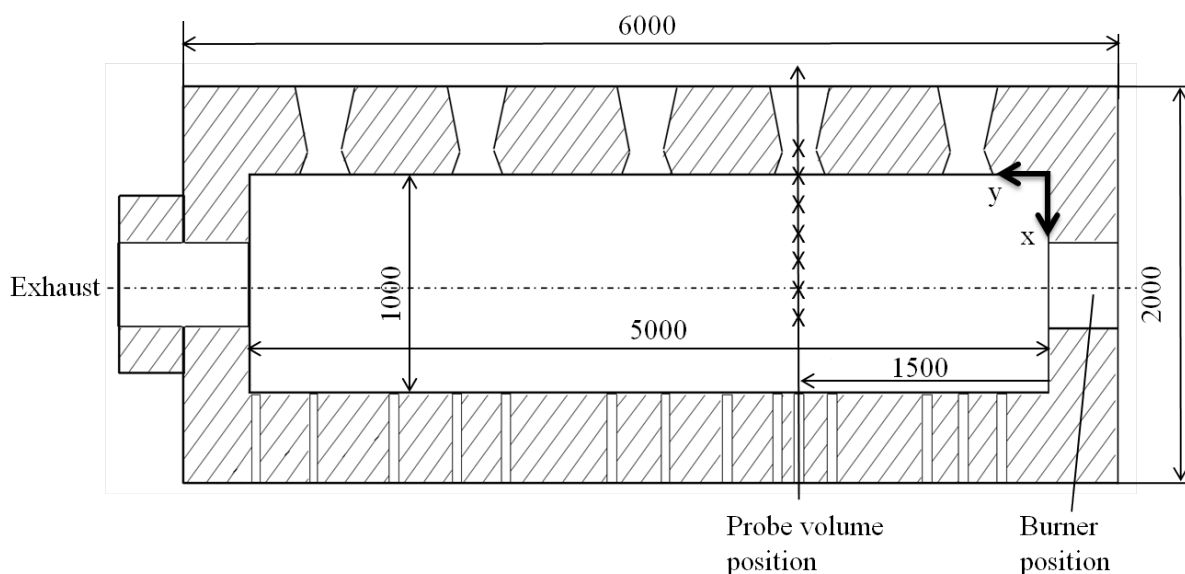

Figure 2:   Sketch of the furnace equipped with the oxy-fuel burner. The probe volume positions of the CARS measurements are marked (x).

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shot to shot variation of the temperature can be observed. At x=270 mm a gas temperature between 727 K and 2697 K and a corresponding large standard deviation of 385 K was measured. Only a small part of the temperature fluctuation is caused by the sensor system. In prior laboratory tests with a McKenna burner in a laminar premixed CO flame a standard deviation of around 100 K at a mean flame temperature of 2000 K was observed. This is typical for a vibrational CARS sensor and comparable values have been found for CARS systems using N_2 as temperature indicator [32].

Nevertheless the achieved gas phase temperatures are lower than the adiabatic flame temperature of 3051 K for the oxy-fuel flame investigated here. This can be explained by additional air sucked into the combustion chamber due to a pressure difference of -0.08 bar to the environment.

As an example in Fig. 4 evaluated single shot spectra are shown together with the best fitting theoretical spectrum. The normalized experimental spectra were compared against a library of theoretically calculated vibrational CARS spectra using a least-squares contour-fitting method. The evaluation procedure interpolates between the precalculated spectra of varying temperatures in the library by means of cubic splines using the Levenberg–Marquardt algorithm. It can be seen from Fig. 4 that the fit quality is quite good for the complete temperature range of interest.

Summary, Conclusion and Outlook

For the first time temporally and spatially resolved O_2 vibrational CARS temperature measurements in a furnace equipped with an industrial relevant oxy-fuel burner were performed successfully. The results show that the developed O_2 vibrational CARS sensor is well suitable for a non-invasive investigation of such combustion processes. A temperature profile along a line for a fixed downstream position was evaluated from
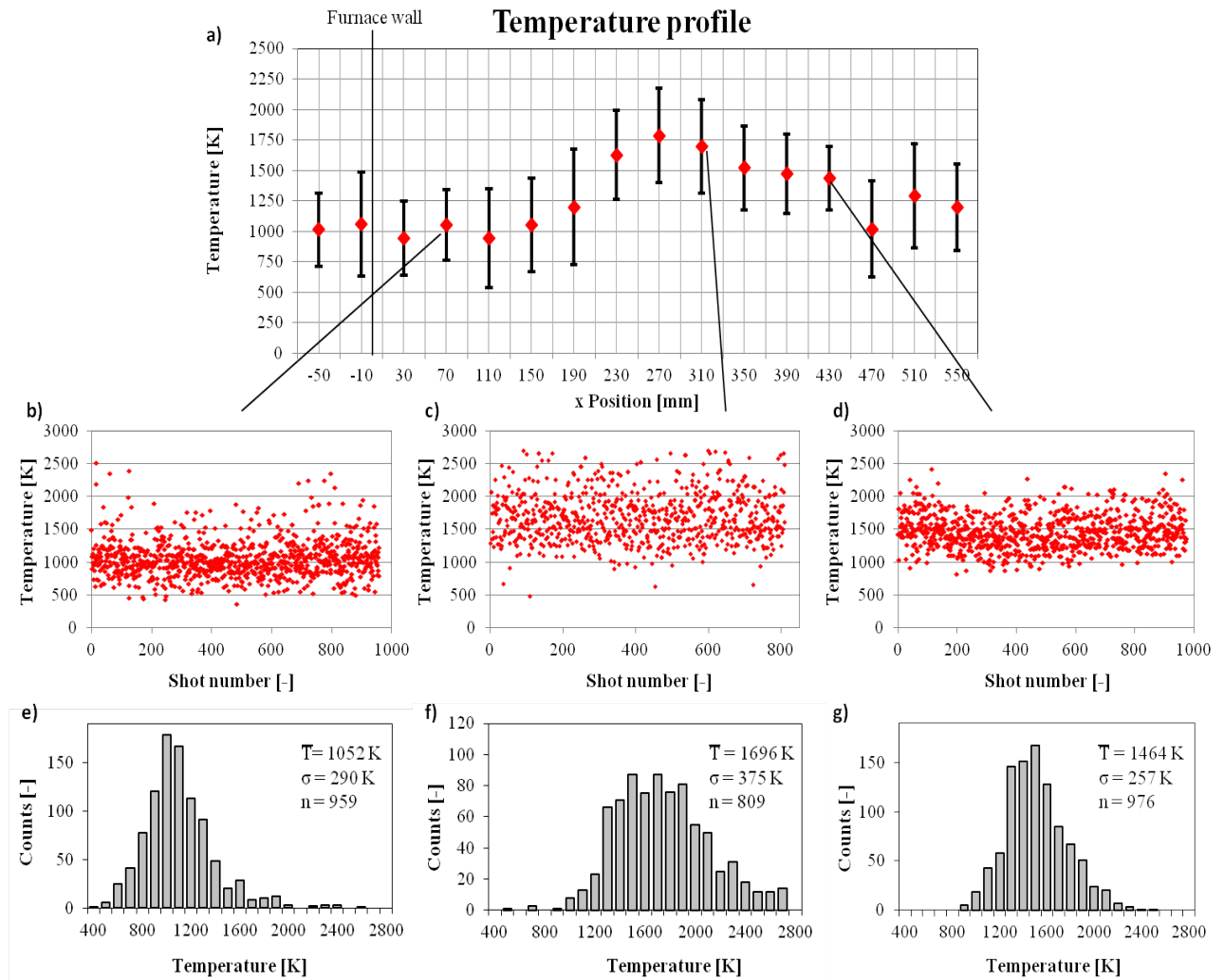

Figure 3: a) Temperature profile taken along a line 1500 mm downstream of the burner position
b-d) Scatter plots for selected measurement positions
e-g) Probably density distributions for the same selected probe volume positions.

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single shot temperature measurements. The corresponding scatter plots show the turbulent behaviour of this oxy-fuel diffusion flame.
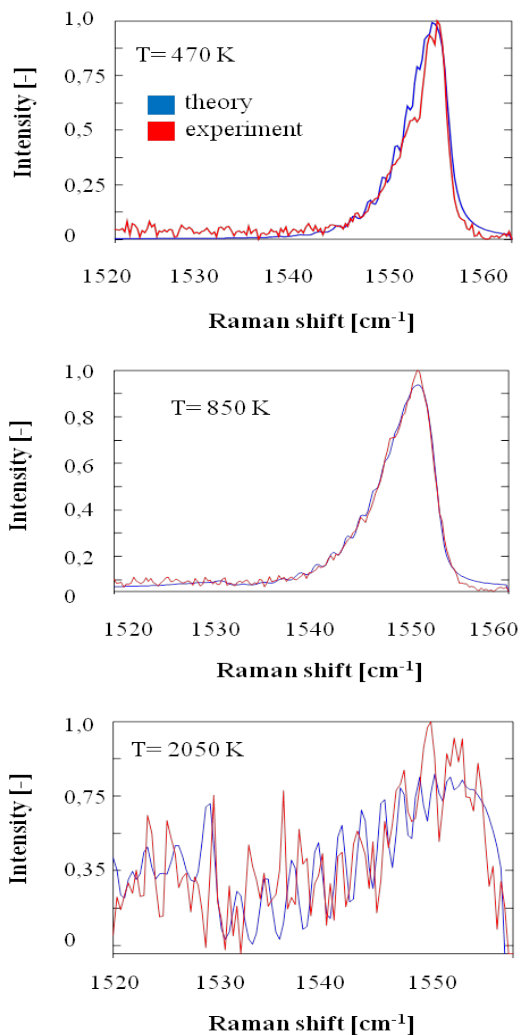

Figure 4.   Selected single shot spectra taken at x=550 mm shown together with the best fitting theoretical spectrum.

Acknowledgement

References

[1]   S. MacLean, J. Leicher, A. Giese, J. Irlenbusch, GWI - Gaswärme International 61, (2012), pp. 85-92.

[2]   R. al-Chawbi, C. Schatz, L. Yap, R. Marshall, Ceramic Engineering and Science 16, (1995), pp. 202-215.

[3]   C. P. Ross, G. L. Tincher, M. Rasmussen, Glass Melting Technology: A Technical and Economic Assessment, Glass Manufacturing Industry/ U.S. Department of Energy-Industrial Technologies Program, USA, #DE-FC-36-021D14315, 2004.

[4]   F. Kluger, P. Mönckert, T. Wild, A. Marquard, A. A. Levasseur, Entwicklungsstand der Oxy-Fuel-Verbrennungstechnologie, in: M. Beckmann, A. Hurtado, Kraftwerkstechnik – Sichere und nachhaltige Energieversorgung, vol. 2, TK-Verlag Karl Thomé-Kozmiensky, Neuruppin, 2010.

[5]   W. Kuckshinrichs, P. Markewitz, J. Linssen, P. Zapp, M. Peters, B. Köhler, T. E. Müller, W. Leitner, Weltweite Innovation bei der Entwicklung von CCS-Technologien und Möglichkeiten der Nutzung des Recyclings von CO_2, Study on behalf of the Federal Ministry of Economy and Energy, Project number 25/08 AZ | D4-020815, 2010.

[6]   J. Warnatz, U. Maas, R. W. Dibble, Combustion: Physical and Chemical Fundamentals, Modeling and Simulation, Experiments, Pollutant Formation, 3rd edition, Springer, Berlin Heidelberg, 2001.

[7]   N. Lallemant, F. Breussin, R. Weber, Analysis of the flame structure, heat transfer and NO_x emission characteristics of oxy-natural gas flames, International Flame Research Foundation, IJmuiden, NL, IFRF Doc No F85/y/7, 1998.

[8]   N. Lallemant, J. Dugué, R. Weber, Analysis of the Experimental Data Collected during the OXYFLAM-1 and OXYFLAM-2 Experiments - Part One, International Flame Research Foundation, IJmuiden, NL, F 85/Y/4, 1997.

[9]   N. Lallemant, J. Dugué, R. Weber, J. Inst. Energy 76, (2003), pp. 38-53.

[10]   A. C. Eckbreth, Laser Diagnostics for Combustion Temperature and Species, 2nd edition, Gordon and Breach, Amsterdam, 1996.

[11]   K. Kohse-Höhinghaus, J. B. Jeffries, Applied Combustion Diagnostics, Tyler & Francis, New York, 2002.

[12]   S. Kampmann, T. Seeger, A. Leipertz, Appl. Opt. 34, (1995), pp. 2780-2786.

[13]   F. Beyrau, A. Datta, T. Seeger, A. Leipertz, J. Raman Spectrosc. 33, (2002), pp. 919-924.

[14]   A. Braeuer, F. Beyrau, T. Seeger, J. Kiefer, A. Leipertz, A. Holzwarth, A. Soika, J. Raman Spectrosc. 37, (2006), pp. 633-640.

[15] P. Magre, F. Aguerre, G. Collin, P. Versaevel, F. Lucas, J. C. Rolon, Exp. Fluids 18, (1995), pp. 376-382.

[16] C. Brackmann, J. Bood, P.-E. Bengtsson, T. Seeger, M. Schenk, A. Leipertz, Appl. Opt. 41, (2002), pp. 564-572.

[17] F. Beyrau, T. Seeger, A. Malarski, A. Leipertz, J. Raman Spectrosc. 34, (2003), pp. 946-951.

[18] A. Datta, F. Beyrau, T. Seeger, A. Leipertz, Combust. Sci. Technol. 176, (2004), pp. 1965-1984.

[19] M. C. Weikl, F. Beyrau, A. Leipertz, Appl. Opt. 45, (2006), pp. 3646-3651.

[20] C. Brackmann, J. Bood, M. Afzelius, P.-E. Bengtsson, Meas. Sci. Technol. 15, (2004), pp. R13-R25.

[21] W. Clauss, D. N. Klimenko, M. Oschwald, K. A. Vereschagin, V. V. Smirnov, O. M. Stelmakh, V. I. Fabelinsky, J. Raman Spectrosc. 33, (2002), pp. 906 – 911.

[22] J. Hussong, R. Lückerath, W. Stricker, X. Bruet, P. Joubert, J. Bonamy, Appl. Phys. B 73, (2001), pp 165-172.

[23] G. Switzer, G. Sturgess, D. Sloan, D. Shouse, Relation of CARS Temperature Fields to Lean Blowout Performance in an Aircraft Gas Turbine Generic Combustor, AIAA Paper 94-3271, 1994.

[24] T. R. Meyer, S. Roy, R. P. Lucht, J. R. Gord, Combust. Flame 142, (2005), pp. 52-61.

[25] T. A. Reichardt, P. E. Schrader, R. L. Farrow, Appl. Opt. 40, (2001), pp. 741-747.

[26] T. Seeger, Moderne Aspekte der linearen und nichtlinearen Raman-Streuung zur Bestimmung thermodynamischer Zustandsgrößen in der Gasphase, Habilitation, University of Erlangen-Nuremberg, 2006.

[27] A. C. Eckbreth, G. M. Dobbs, J. H. Stufflebeam, P. A. Tellex, Appl. Opt. 23, (1984), pp. 1328-1339.

[28] E. Magens, Nutzung von Rotations-CARS zur Temperatur- und Konzentrationsmessung in Flammen, Dissertation, University of Erlangen-Nuremberg, 1993.

[29] L. A. Rahn, R. E. Palmer, M. L. Koszykowski, D. A. Greenhalgh, Chem. Phys. Lett. 133, (1987), pp. 513–516.

[30] K. P. Huber, G. Herzberg, Molecular Spectra and Molecular Structure, vol. 4, Van Nostrand, Reinhold, New York, 1979.

[31] G. Rouille, G. Millot, R. Saint-Loup, H. Berger, J. Mol. Spectrosc. 154, (1992), pp. 372-382.

[32] A. Thumann, T. Seeger, A. Leipertz, Appl. Opt. 34, (1995), pp. 3313-3317.