EVALUATION OF OPTICAL AND SPECTROSCOPIC EXPERIMENTS OF HYDROGEN JET FIRES

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ABSTRACT

This paper reports results of evaluating joint experiments under the work programme of Hysafe occurring at HSL who provided the test facilities and basic measurements to generate jet fires, whereas Fraunhofer ICT applied their equipment to visualise the jet fires by fast video techniques, IR-cameras and fast scanning spectroscopy in the NIR/IR spectral region. Another paper describes the experimental set up and main findings of flame structures and propagation resolved in time. The spatial distribution of species and temperate as well as their time history and fluctuations give a basis of the evaluation of effects caused by such jet fires. Fraunhofer ICT applied their comprehensive evaluation codes to model the radiation emission from 3-atomic species in the flame, especially H_2O in the Infrared spectral range. The temperatures of the hydrogen flame were about 2000 K as found by least squares fit of the measured molecular bands by the codes. In comparison with video and thermo camera frames these might enable to estimate on a qualitative level species distribution and air entrainment and temperatures to identify hot and reactive zones. The risk analysis could use this information to estimate heat transfer and the areas of risk to direct inflammation from the jet fires by semi-empirical approaches.

1.0 INTRODUCTION

Many chemical reactions in industrial processes use hydrocarbons or hydrogen in pressurized closed vessels. On accidents these might release hydrogen to generate hot flame jets on ignition. Depending on the case of leakages these gases may be mixed with air or oxygen so they might ignite, burn, explode or even detonate. Gas explosions in closed vessels produce high pressure forces on the complete enclosure which can lead to total damage of a reactor or at least to leakages to release the complete stored mass of gas. Radiation and direct contact might initiate fires and explosions. The explosive reactions occur in wide ranges of fuel/oxidizer compositions with flame velocities up to more than 100 meter per second, which are strongly enhanced by turbulence or in spherical explosions [1 - 5]. Pressure waves dominate the effects of gas explosions. Radiation of the hot species depending on temperature is the main effect to influence the surroundings and contribute to fire and explosion transfer and propagation by its strong heat transfer. Especially, the dominating risks of hydrogen jet fires emerge from direct contact of the flame and its reacting species, the final combustion products and the dispersion of hot gases. The radiative emission of hazardous fires and gas/liquid explosions is strongly variable in time by jets lasting till shut down or complete release and at short scales down to milliseconds [6]. In addition, it includes fundamental information concerning the mechanisms and progress of burning. As investigated in the described experiments, a fully deployed hydrogen jet (un-reacted) generates an explosion when ignited at a point close to stoichiometry and transfer to a turbulent flame jet expanding the volume of the original jet. A scanning analysis of the spectral flame radiation must record the flame dimensions, be fast enough to detect pre-reactions, transient phenomena, starting explosions, enable the understanding of propagation mechanisms as well as quenching or extinguishing mechanisms. In contrast to pressure effects, time and spectrally resolved radiation of fires and explosions is not investigated in a sufficient way, especially concerning spectral resolution and influence on environment and there exist only limited publications on measurements resolved in time and wavelength [6 - 12].

The results reported in this paper were generated by evaluating joint experiments under the work programme of Hysafe occurring at HSL (Buxton) in 2008. Basic measurement and control were done by HSL at their test facilities to generate jet fires. Fraunhofer ICT used their equipment to visualise the jet fires by fast video techniques, IR-cameras and fast scanning spectroscopy in the NIR/IR spectral region. The results presented here focus on spectroscopic measurements in the NIR/IR spectral region and include only a partial evaluation because the high amount of information from 23 jet experiments generated by the various optical and spectroscopic equipment applied, all applied in high time resolution of about 10 ms. This paper should be read together with the paper "Visualisation of Jet Fires from Hydrogen Release**"** also published in this conference proceedings which describes in more detail the shape and structure of the jets by imaging techniques.

2.0 EXPERIMENTAL SETUP

The experimental campaign at HSL investigated hydrogen jets with various mass flow rates. Hydrogen was supplied from a tank which was pressurised by 20MPa and released to the free air restricted by orifices varying in diameter from 1.5 to 3.2, 6.35 and 10 mm in a horizontal exit. An electrical igniter for a jet engine and in some cases a squib initiated at a point located 2.2 or 2.5 m downstream the jet flow. The applied Fraunhofer equipment consisted of high-speed-video, an IR-camera and a spectral radiometer for the IR-range between 1.6 and 14.5 microns with maximal scan rates of 100 scans / s. The spectral radiometer observed one isolated spot of size 0.2 m^2. The spectral radiometer and the IR-camera had a distance of 38.5 m at an aspect angle of about 80° observing the spot at 160 cm and 400 cm from the hydrogen exit. The setup is shown in a sketch in figure 1. Details of the experiments are given in the other paper "Visualisation of Jet Fires from Hydrogen Release**".**

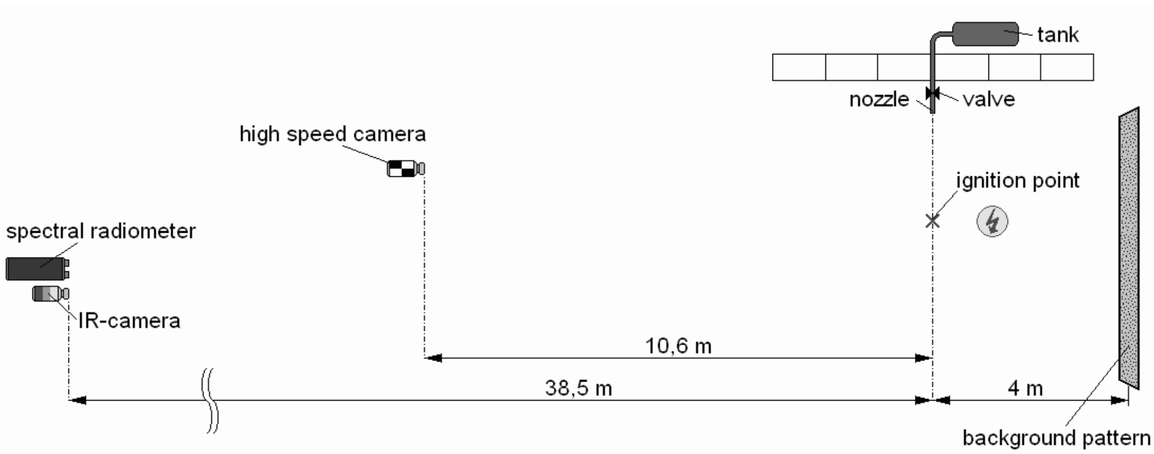


Figure 1: Sketch of the experimental setup

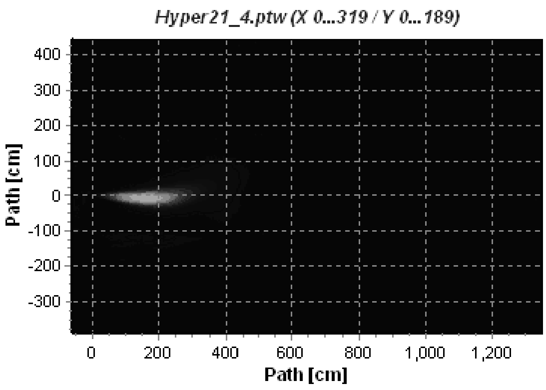
3.0 METHODS OF SPECTRSOCOPIC DATA ANALYSIS

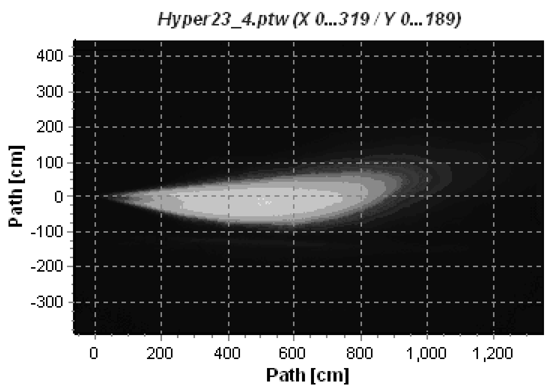
The spatial distribution of species and temperate as well as their time history and fluctuations give a basis of the evaluation to the main risks of such jet fires. The hydrogen jet and especially the flame are not directly visible in the visual spectral range. The flame emits only in the UV, the OH bands located with the strongest band at 306 nm and various series of strong water bands in the NIR and IR spectral regions [6, 8-10]. In an earlier paper the principles were described in more detail applying the technique to hydrogen problems [11, 12]. A quantitative spectral analysis of the measured infrared spectra from flames and plumes of jets has to use molecular band modelling of the species involved. A specific code BAM was developed at Fraunhofer ICT which calculates NIR/IR-spectra and enables a least squares fit to experimental spectra with temperature and concentrations being the fitting parameters (see the summarizing reference [13]). The code utilises the data from the Handbook of Infrared Radiation of Combustion Gases [14, 15] which cover the temperature interval from 600 to 3000 K. The BAM code can calculate NIR/IR-spectra (1-10 µm) of gas mixtures of H_2O (with bands around 1.3, 1.8, 2.7 and 6.2 µm), CO_2 (with bands around 2.7 and 4.3 µm), CO (4.65 µm), NO (5.3 µm) and HCl (3.5 µm). It can take into account emission of soot particles as well as self absorption and pressure line broadening. The spectral bands of three-atomic molecules consist of thousands of single lines, e.g. HITRAN [16, 17] lists nearly 50,000 lines for H_2O and 60,000 for CO_2 and simplified models to quickly obtain line positions and strength are currently not available. The HITRAN code was designed for atmospheric absorption which is here also used to correct the recorded spectra to take into account the distance between experiment and radiometer. Such an IR-radiometer and the data evaluation were already applied to transient large hydrogen jet flames from 75 l hydrogen tanks [10]. Recent published results [19, 20] are based on calculations and similarity considerations using the RADCAL code [21]. This code is based on the same principles like the BAM code, but does not enable least squares fit procedures of spectral to obtain temperatures and species.

4.0 TIME RESOLVED IR SPECTRA

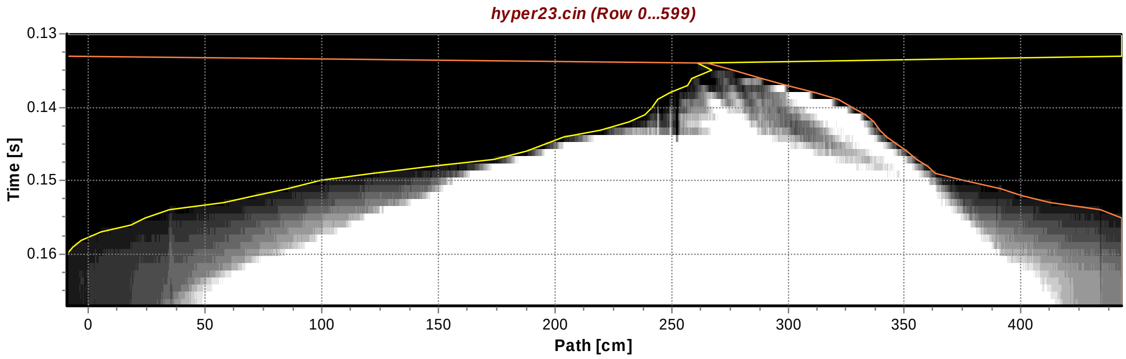
Results of the investigations to visualise the transient flame jet contours applying various methods of video techniques of image analysis are reported in the already mentioned separate paper “Visualisation of Jet Fires from Hydrogen Release**”**. Behind the hydrogen exit a jet forms with a cone half angle between 8-10^{o}. The flame starts after some distances from the exit, being enlarged by a factor of 2-3 compared to the unreacted jet. After reaching a maximum the hot combustion products of the jet remain stable and then cool down. However, also in this latter phase they might endanger the environment as the emitted radiation is quite high for some seconds. However, an unconfined hot gas volume expands, rises because of buoyancy and cools down earlier shown in fig.2b.

The mass flow defines the shape and size of the jet and the burning volume of it (fig. 2). These data are reported in the separate paper analysing the optical structure of the jet. Fig. 2 shows two examples of the averaged flame contours. The spots observed by the radiometers are covered by the flame shortly after ignition and then throughout of the experiment. The diameters of the luminous jet contours at the measuring spots of the radiometer are 50 cm, 100 cm, 140 cm and 140 cm for the orifice diameters of 1.5, 3.2, 6.35 and 10 mm, however fluctuations strongly modulate these diameters by approximately ± 20%. Due to the nearly constant cone angle of the jets the diameters of the longer flames are similar at the observed spot. According to the simulation presented earlier [11], at these a diameters the flames are optically thin at least for the band below the wavelength of 5µm. The measured radiation intensities and relative species concentrations, especially water from the combustion and CO_2 from entrained air can only be quantitatively used for the conditions at that point. The evaluation of the spectra by the BAM code can only be applied to the temperature including a concentration*length. Therefore also ratios of concentrations like that of water and CO_2 can be discussed with reliability. Additional evaluation in the future, using the detailed contours and the BAM code will enable an estimate of the total quantitative radiation resolved wavelength emitted. This evaluation has to take into account both wavelength of radiation and diameter of the jet, because the different bands show different emissivities [13-17].





(a) (b)



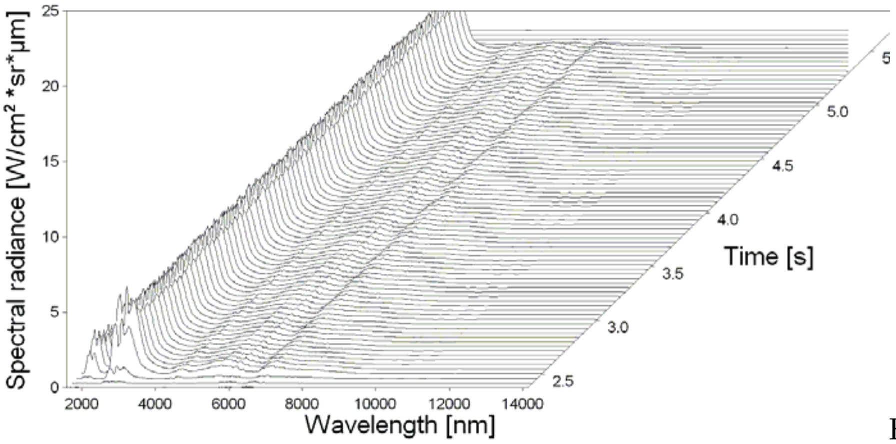
(c)

Figure 2: Averaged flame contours from (1.5 mm orifice diameter Hyper21 and 10 mm orifice - 23) (a) with orifice diameter of 1.5 mm (b) with orifice diameter of 10 mm (c) flame expansion starting from the ignition point from the exit upstream (left) and down-stream (right), (10 mm orifice diameter Hyper23)

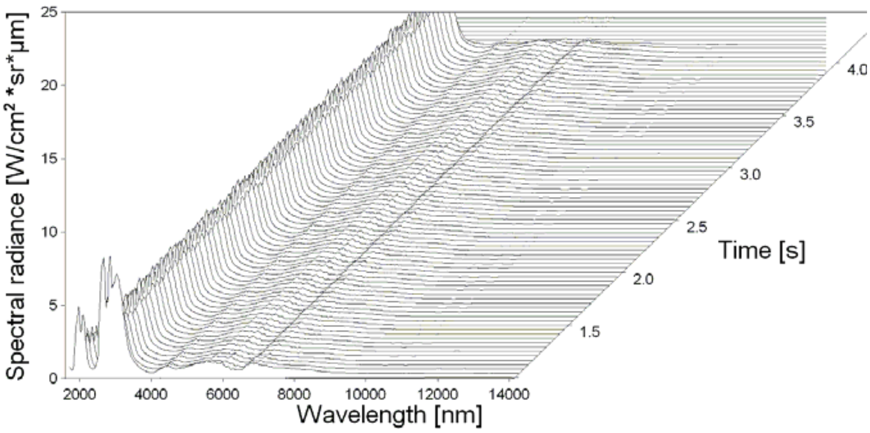
Examples of a series of spectra in the Infrared spectral region are shown in fig. 3. Immediately after ignition the reacting jet emits highest intensities which correspond to the expanding explosion which then relaxes to turbulent jet burning. According to the diameter of the jet at the measuring spot the intensity increases, especially at the phase of the initial explosion (see mentioned paper on visulation). At 160 cm after the hydrogen exit the stable combustion phase leads to similar intensities of the jets from 1.5 mm orifice and that from the 6.35 mm orifice (compare 02 and 07 in fig. 3). At an increased jet diameter the intensity rises as well (compare 07 and 08 in fig. 3).

5.0 SPECTRA EVALUATION

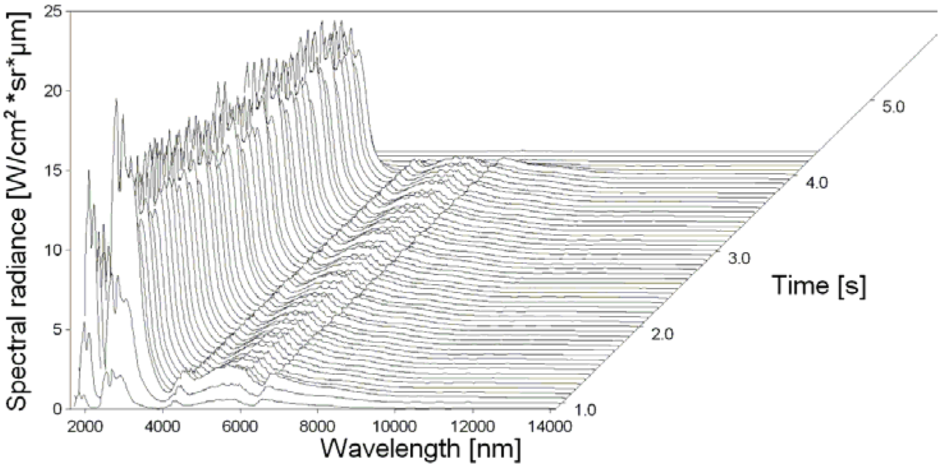
One spectrum is extracted and shown in detail in fig. 4 where, in addition, a calculated spectrum is plotted which corresponds to the best fit with a temperature of about 2300 K. It contains all the well known water bands and a weak carbon dioxide emission at 4.25 µm which is entrained by air. The main differences between the measured and calculated spectrum occur at the wavelength positions of CO_2 at 2.7 µm and 4.25 µm where possibly self absorption and atmospheric absorption is not yet correctly included. The other spectral areas are in close agreement.



Hyper02



Hyper07



Hyper08

Figure 3: Series of IR spectra on the progress of the jet combustion, (Hyper 02 measurement spot at 160 cm, -07 at 160 cm and -08 at 400 cm)

Fig. 5 shows the temperature of the measured spot depending time for the series Hyper01. The temperatures are for this experiment all above 2300 K below the adiabatic flame temperature of stoichiometric hydrogen air flames [18]. Due to the low mass flow rates and fluctuation are not occur substantially with respect to temperature.

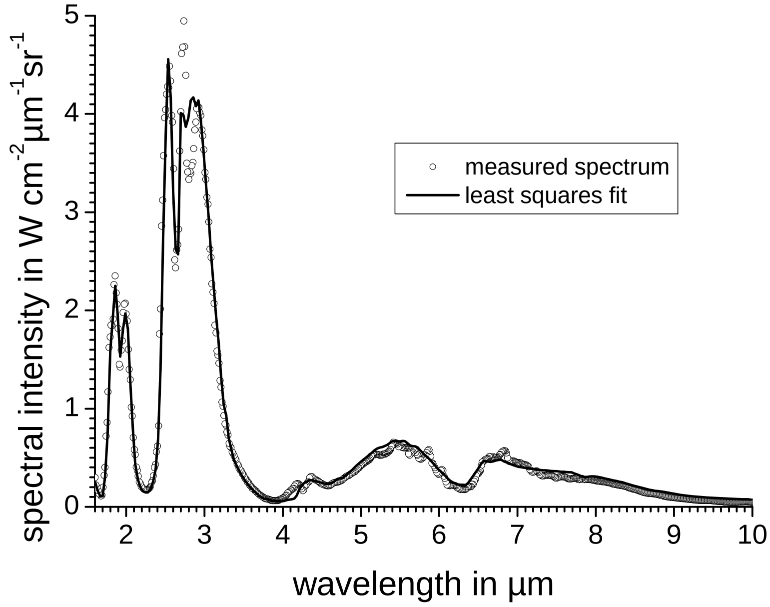


Figure 4: Extracted spectrum from the series of Hyper01 and the least squares fit by the BAM code, the weak CO_2 band (at 4.25 µm) can be also used by the fit

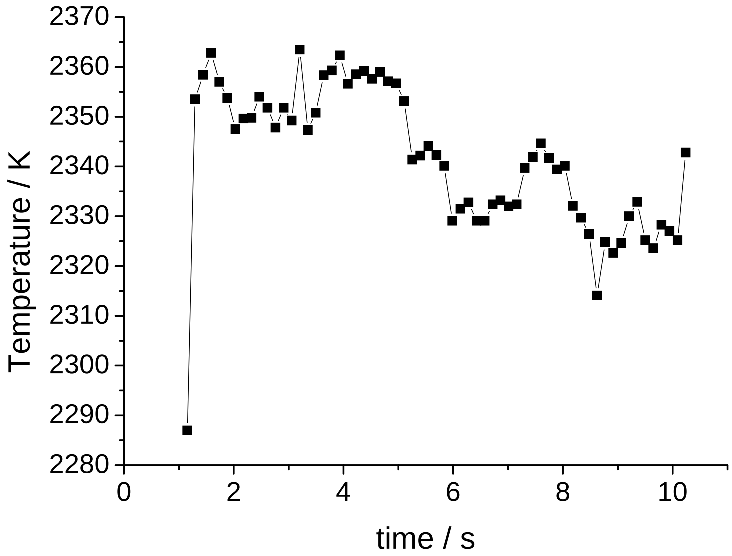


Figure 5: Temperatures from the series of Hyper01 and the least squares fit by the BAM code

The obtained temperatures of all 23 experiments and the individual spectra are plotted in fig 6. The BAM calculations give also species concentrations*pathlength, beneath the temperatures. Although the jet thickness is known it might be inhomogeneous. Therefore ratios of these data can be used without more detailed analysis which will be done in a future paper. It is interesting to study the ratio of H_2O to CO_2 as the latter can be only entrained by air in the turbulent jet flow. Assuming that CO_2 would not decompose it is the residue of the 0.058% contained in air. In fig. 5 these ratios are correlated with the temperature.

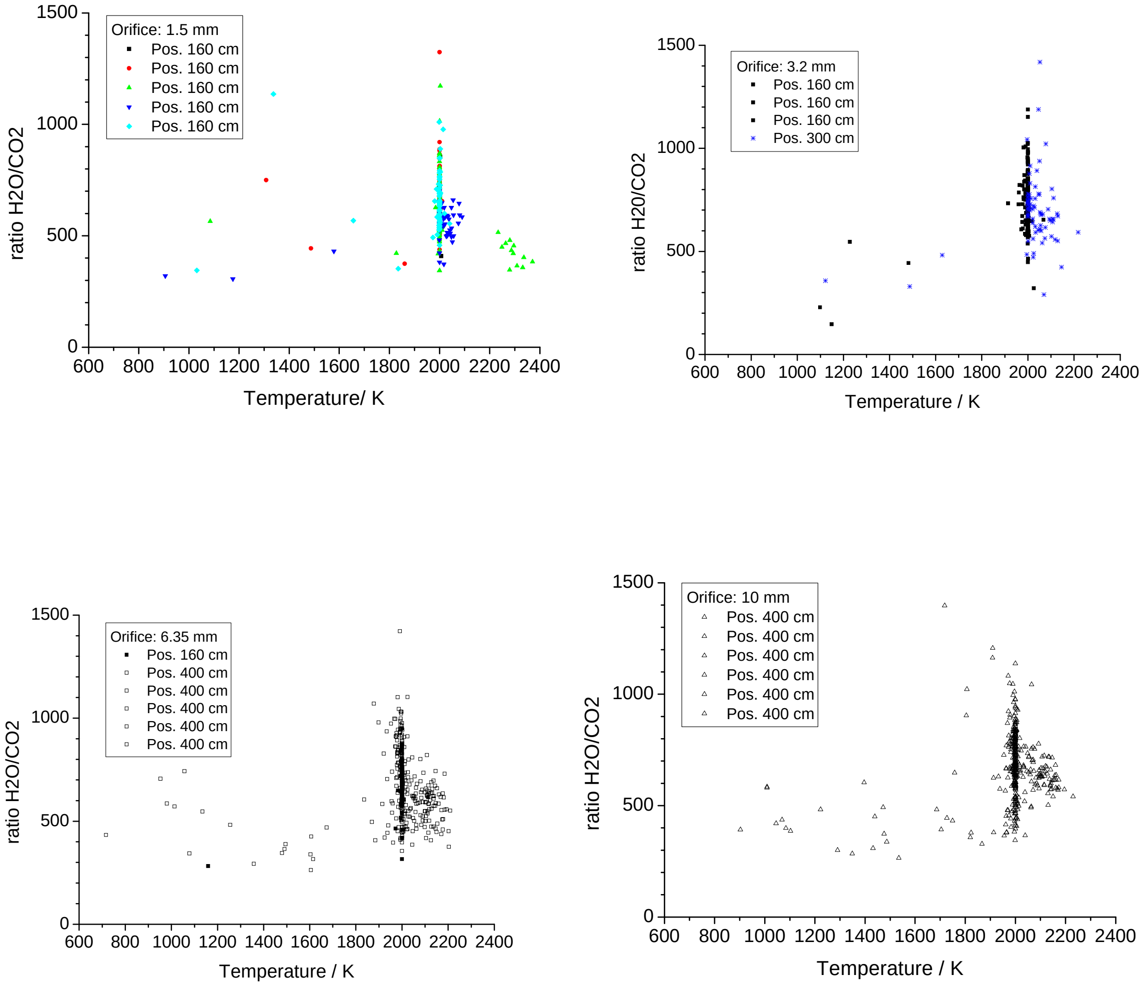


Figure 6: Temperatures and ratios of water to carbon dioxide for all experimental series, obtained by application of the least squares fit by the BAM code,

The temperatures from all 23 experimental and for each individual spectrum of each experiment obtained with a time resolution of 10 ms accumulate at 2000 K for all spectra. This means that at the measurement spot in about 80-90% of the measurement time interval a temperature close to 2000 K is found. The ratio of water to CO_2 remains between 300 and 1000 which means at the measuring spots the air content was always higher than needed for a stoichiometric mixture.

There are clusters (in total approximately 10%) ranging from 2000 to 2400 K, in addition. This might be caused by passing of turbulence structures where hydrogen was burnt by air in stoichimetric conditions. A scatter of some temperature values range from 700 to 2000 K. These values may be attributed to structures of fresh air entrainment by turbulence.

By thermodynamic calculations these ratios can be calculated in dependence of the temperature and water concentrations in the flame. Figure 6 shows results of using the ICT code [18] varying CO_2 concentrations in air. It relates the ratios of water to carbon dioxide in the flame to the water concentration and the temperatures in the flame. The highlighted curves concern the normal CO_2 concentration in air and suggest the surrounding area to be the most probable situation of the experiments. These are marked by the framed areas in the diagrams corresponding to the results in fig. 7. This means that the combustion takes place at the measured spots to generate temperatures of about 2000 K and water concentrations (neglecting fluctuations) between 0.17 and 0.25. This corresponds to an air content in the mixture beyond stoichimetry before reacting confirming the situation as already mentioned above.

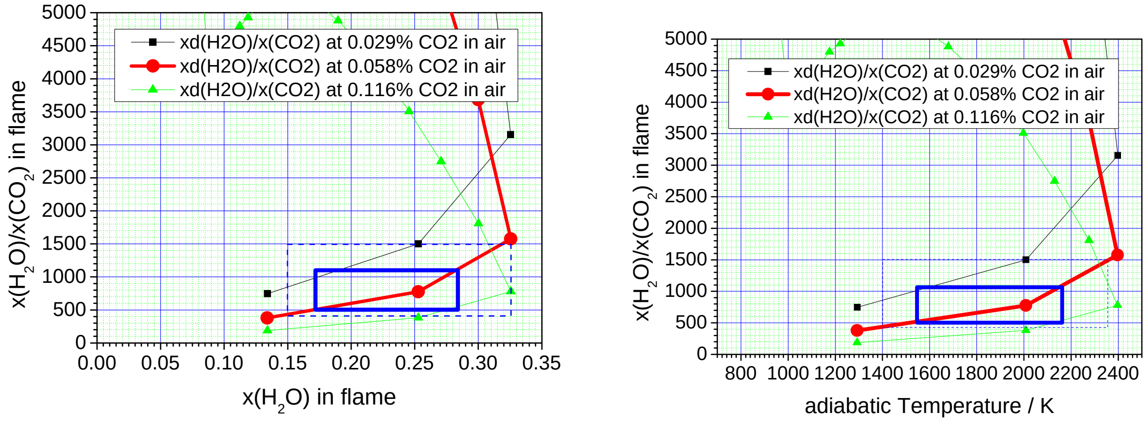


Figure 7: Ratios of water to carbon dioxide in the flame related to the water concentration and the temperatures in the flame; the highlighted curve concerns the normal CO_2 concentration in air; the framed area in the diagrams are those corresponding to the results in fig. 5.

The evaluation of the results will be continued and a more detailed analysis of the results published in a future paper. Especially correlations with overall simulation approaches [19, 20] will be considered if further details are extracted from the data.

6.0 CONCLUSION

A series of experiments of hydrogen jets were investigated with flow rates of 40 to 670 g/s with flame lengths from 3 to 10 m. The analysis of the spectroscopic results by modelling the bands of water and carbon dioxide result in the following conclusions:

- The temperatures of a flame jet spot accumulate at 2000 K reaching to 2400 K, some aspects of the turbulence structure are evident however, need further evaluation
- Fast scanning spectrometers and the BAM code enables a detailed analysis of a spot currently, and need imaging spectrometers to cover a full jet simultaneously which might verify the modelling approaches by radiation codes.
- A quantitative estimation of the total emitted radiation is only possible by a correlation of the flame contours assuming air entrainment according to the jet expansion on its length by using the BAM code. This evaluation is planned for the future.

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