

# Chirped-probe-pulse femtosecond coherent anti-Stokes Raman scattering for gas-phase temperature measurements in high-pressure kerosene/air flames

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## ABSTRACT

Air traffic is increasing and its related emission is a major concern for the stability of the planet's climate. Innovative combustion systems need to be developed to continue to enable mobility while respecting the environment. The qualification of new propulsive systems requires the analysis of the associated combustion and one of the quantities of major interest is the temperature. The present work exposes the development of the chirped-probe-pulse femtosecond coherent anti-Stokes Raman scattering (CPP-fs-CARS) thermometry at 1 kHz and its application in a semi-industrial test bench.

CARS is a third order nonlinear optical diagnostic renowned for high accuracy temperature determination. CPP-fs-CARS rely on the analysis of the frequency-spread dephasing rate after the initial excitation of the Raman coherence on  $N_2$ . This pump-probe method needs three input pulses. The two first excitation pulses are Fourier transform limited and present a temporal width of 100 fs at 800 nm and 675 nm. The frequency difference matches  $N_2$  molecular vibrational energy gap. The evolution of the coherence generated in the medium is probed with a delayed picosecond which encompasses a sufficient part of the coherence evolution. In the CPP configuration, this longer probe pulse results from a 100 fs, 675 nm temporally stretched through a propagation in a 30 cm glass rod. Following the energy conservation principle, the interaction results in the generation of a CARS signal, presenting a spectral shape sensitive to temperature. Temperature is deduced from the single-shot CARS spectra adopting a genetic algorithm. First results will report the application of the diagnostic in atmospheric pressure well known environments and the associated data processing. Such environments lead to found a temperature accuracy in a near-adiabatic laminar flame better than 1.5% at 2250 K.

Those results demonstrate the applicability and usefulness of CPP-fs CARS and are pursued with the investigation of a two-phase flames at a pressure of 0.75 MPa representative of aircraft combustor operating point. Measurements were performed at several locations in planes orthogonal to the flame propagation and at several distances from the injection system. The data processing of the measurements in such harsh conditions. Finally, temperature evolution was extracted from single shot measurements and probability density function (PDF) are reported at several locations of interest enabling to retrieve not only the mean temperature, but also important information on flame behaviour at different stages of its evolution within the combustion chamber length.

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## 1. Introduction

For nearly four decades, coherent anti-Stokes Raman scattering (CARS) spectroscopy has been used to measure both temperature and species concentrations of major species in combustion environments (A.C.Eckbreth, 1996) (Druet & Taran, 1981). CARS spectroscopy is well suited for temperature and species measurements due to its direct measurement of the temperature dependent Boltzmann-distributed population of vibrational and rotational energy states. In this regard, the spectral profile of the CARS signal is highly temperature dependent, while the amplitude of the signal is related to the concentration of the targeted species. In the case of experiments in air-breathing combustors,  $N_2$  is frequently used as the probe molecule due to its low reactivity and high relative concentration everywhere in the flow. Furthermore, CARS is particularly applicable to high-temperature and high-pressure environments where others optical (laser-induced fluorescence, Raman and Rayleigh scattering...) and non-optical diagnostics (thermocouples...) present large limitations.

As a laser-based diagnostic tool, in-situ temperature measurements are possible, meaning that the laser/molecules CARS interaction is non-invasive and occurs only in a small sample volume. Additionally, the CARS signal exhibits large SNR due to its third-order dependence on incident laser power, and this one can be spatially separated from background scatter with high collection efficiency. As a result, CARS can be applied in harsh operating conditions where others measurement techniques are not applicable. However, several challenges complicate the CARS data reduction and limit accuracy and precision in practical combustion systems. For instance, turbulent combustion systems relevant to gas turbines typically operate at pressures up to 6.0 MPa and may exhibit significant pressure fluctuations both during normal and unsteady operation. Until now, four main challenges have driven innovation in gas-phase CARS research : the simultaneous measurement of temperature and concentration of several species using a single CARS system, the application of CARS to challenging combustion environments such as high-pressure two-phase flames, the suppression of non-resonant background and collisional energy transfer effects in CARS measurements at high pressure and high temperature, and the measurement of temperature and species concentration at high repetition rates (1 kHz or higher) to investigate transient instabilities and phenomena associated with turbulent reacting flows.

Although many innovative adaptations on CARS have been mainly employed in the ns temporal regime, no single CARS configuration can meet all these challenges. For instance, the primary advantage of ns-CARS was the ability to spectrally resolve closely-spaced Raman transitions with high pulse energy. Thus, time-domain information was ignored in favor of detailed spectral

information. While the ns-CARS has proven its potential in harsh conditions (Grisch, Bouchardy, & Clauss, 2003) (Roy, Gord, & Patnaik, 2010), the lack of high-repetition-rate laser sources and existence of composition-dependent contributions in complex flows are still limiting points. These reasons have then motivated a new strategy requiring ultra-short laser pulses in the femtosecond regime. Indeed, since the early 2000s, a new interest for CARS has arisen with the enhanced performances of femtosecond laser commercial sources. The main advantage is that single-shot temperature CARS measurements can be theoretically performed at data rates up to 10 kHz, thousand times larger than in the nanosecond regime offering a way to study dynamic processes as well as a reduction in the duration of the experiments. Two emerging experimental methods suitable to perform single laser-shot fs CARS temperature measurements have been developed and applied in recent years, these ones being referred to the hybrid fs/ps CARS and CPP-fs CARS (Richardson, Lucht, Kulatilaka, Roy, & Gord, 2013) (Miller, Dedic, & Meyer, 2015) (Kearney & Scoglietti, 2013). In the current study, CPP-fs CARS was selected because temperature measurements can be performed from the frequency-spread dephasing rate after excitation of the Raman coherence. By employing a chirped-probe-pulse (CPP) strategy, the Raman coherence decay can be probed with a single-laser-shot in the frequency domain. Furthermore, CPP-fs CARS has the potential to deliver temperature measurements with greater accuracy in comparison with traditional ns-CARS because molecular collisions do not affect the CARS process, that is, Raman linewidths need not be determined for CPP N<sub>2</sub> fs-CARS thermometry at least for atmospheric pressure (Lucht, Roy, Meyer, & Gord, 2006). Finally, the CPP fs-CARS technique allows for suppression of the non-resonant background signal by delaying the probe beam in time without losing temperature measurement accuracy.

The current paper focuses on high-repetition rate, high-dynamic range temperature measurements using N<sub>2</sub> CPP-fs CARS in high-pressure kerosene/air flames. The next section describes essential features on the CPP-fs CARS system developed for this purpose. Then, the experimental procedure for the data reduction of CARS spectra is outlined. Finally, the first part of the result section is dedicated to the description of CARS measurements in low and high-temperature flows used to determine the temperature accuracy while the second part examines the applicability of the CPP-fs CARS for measuring temperature at high-repetition rate in high-pressure kerosene/air flames.

## 2. Experimental Apparatus

Experiments were conducted with a Ti:Sapphire femtosecond laser system consisting of an oscillator (Coherent Vitesse) combined to an amplifier (Coherent Legend Elite Duo HE + III) which delivered laser pulses of  $\sim 95$  fs (FWHM) duration at 800 nm at a repetition rate of 1 kHz. The oscillator source provided short pulses of few nJ at a frequency rate of 80 MHz. Amplification was performed by a CPA (Chirped Pulse Amplification) system consisted of a stretcher which temporally expands the pulse and reduce its peak power, followed by two amplification stages. The first amplification stage consisted of a multi-passage cavity optically pumped by a 23 W laser source (Coherent Evo 45) to deliver an energy of  $\sim 7.5$  mJ. The second stage was optically pumped with a counter-propagative process by a second 45 W laser source (Coherent Evo HE) to reach in a single pass pulse energy of  $\sim 15.8$  mJ. After a temporal recompression, the resulting energy of the laser pulse was  $\sim 12$  mJ. The output laser beam was then split so that half of the energy of the 800 nm laser beam pumped an Optical Parametric Amplifier (OPA, TOPAS Prime Plus) while the second part was used as the Stokes beam. The output beam from the OPA was at 675 nm and the energy was set to  $520 \mu\text{J}$ . The resulting OPA beam was subsequently divided into two parts for producing the pump and the probe beams. The probe beam was chirped by inserting a 30 cm long SF11 glass rod into the beam path for stretching the temporal width of the pulse from 95 fs to 4.3 ps. The temporal coincidence of the pulses at the probe volume was controlled with two motorized linear translation stages installed along the beam path of the pump and the probe beam. Control of energies of the laser pulses at the exit of the CARS setup was adjusted by using neutral density filters and thin-film polarizer and half-wave plates. The laser beams were then arranged in a folded BOXCARS configuration and focused into the measurement probe of 1.2 mm long and  $50 \mu\text{m}$  in diameter using a 250 mm focal length lens. The energies deposited into the probe volume for the pump, Stokes and probe pulses were  $35 \mu\text{J}$ ,  $35 \mu\text{J}$  and  $210 \mu\text{J}$  respectively. These energies enable CARS measurements without laser pulse spectral distortion which can be induced by the density of the medium. After collimating the CARS signal with a 250 mm focal length lens, this one was spatially and spectrally filtered from the incident laser beams and then transported by mirrors to be focused to the entrance slit of a 0.32 m spectrograph (Princeton Isoplan 320) equipped with a 1200 lines/mm grating. An emCCD camera with a  $512 \times 512$  pixels triggered by the fs-laser time circuit, completed the detection system and assumed the acquisition of the CARS spectra at a repetition rate of 1 kHz. It should also be noted that part of the optical line was also fixed on a

2D translation stage in order to perform CARS temperature measurements at different positions of the high-pressure flame.

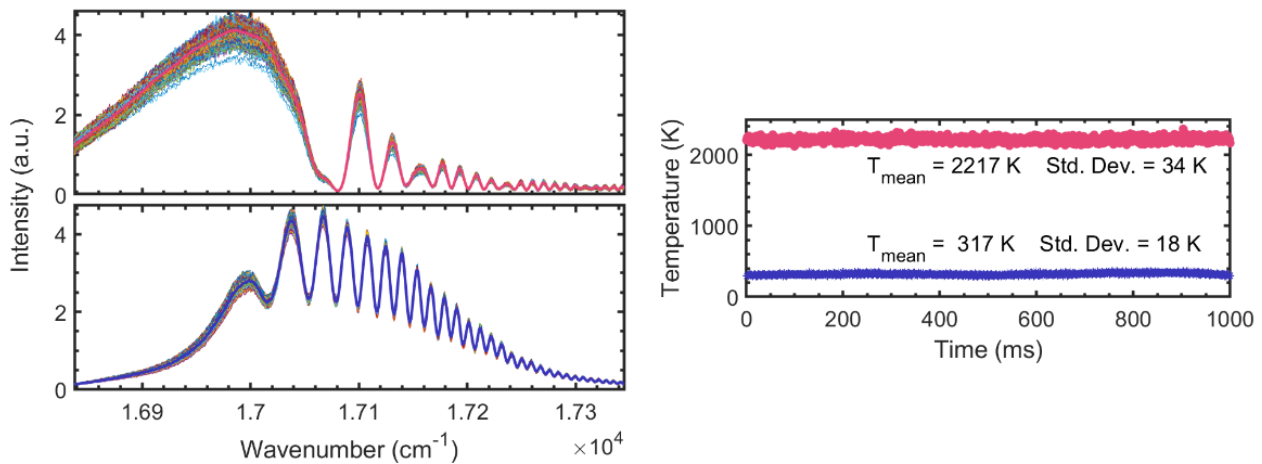
### 3. CARS data reduction

The CPP fs-CARS diagnostic has been specifically developed to perform temperature measurements at high repetition rates in high-pressure reactive flows. After recording the CARS spectra into the flame, a CARS data reduction procedure was performed using an in-house written software. The first step of the data reduction of CARS spectra was to subtract background emission. The background emission was either collected in the emCCD camera when no CARS signal was produced. The experimental CARS spectra were then compared to numerical simulations using a "Differential Evolution" genetic algorithm [5] which aims at minimizing by a least-squared method, the difference between the single-shot CARS spectrum and the theoretical spectrum presenting the best agreement. An iterative procedure is then set up to identify the optimal set of parameters presenting the best minimization between the experimental and theoretical spectral shapes of the CARS signals. In this procedure, the parameters to be adjusted by the genetic algorithm are twelve: nine of them are related to the laser parameters such as the 2<sup>nd</sup> and 3<sup>rd</sup> order spectral phase parameters of the Pump, Stokes and probe laser beams, the relative delays between laser pulses and the spectrum of the three laser pulses while the other ones are the ratio  $\beta/\alpha$  and the phase shift  $\varphi$  between the resonant and non-resonant contributions and the temperature. Values of the nine laser parameters are determined by fitting a reference calibration CARS spectrum recorded at known temperature, species composition and pressure. These laser parameters are then kept constant when the CARS spectra recorded at unknown thermodynamic conditions are compared with theoretical spectra. In this case, only the three other parameters remain free.

### 4. Results

To validate the data reduction procedure, N<sub>2</sub> single-shot CARS spectra were first recorded in an air jet heated at 45°C, then in a stoichiometric laminar premixed CH<sub>4</sub>-air flame. An example of results obtained during these experiments is depicted on Fig. 1. For each operating condition, the mean experimental CARS spectrum is superimposed to 1000 single-shot CARS spectra. First, it is remarkable to note the reproducibility of the single-shot CARS spectra which is mainly related to the good temporal stability of the laser energy pulses emitted by the femtosecond laser. Thus, the

spectra recorded in these conditions allow to measure the mean temperature and rms temperature fluctuations and an accuracy better than 6 % at 317 K and 1.5 % at 2240 K is obtained.



**Fig. 1:** 1000 instantaneous and resultant averaged CPP fs-CARS spectra recorded at 1 kHz in a heated air flow and a CH<sub>4</sub>/air premixed flame (left) and associated time history profiles of single-shot temperature measurements (right)

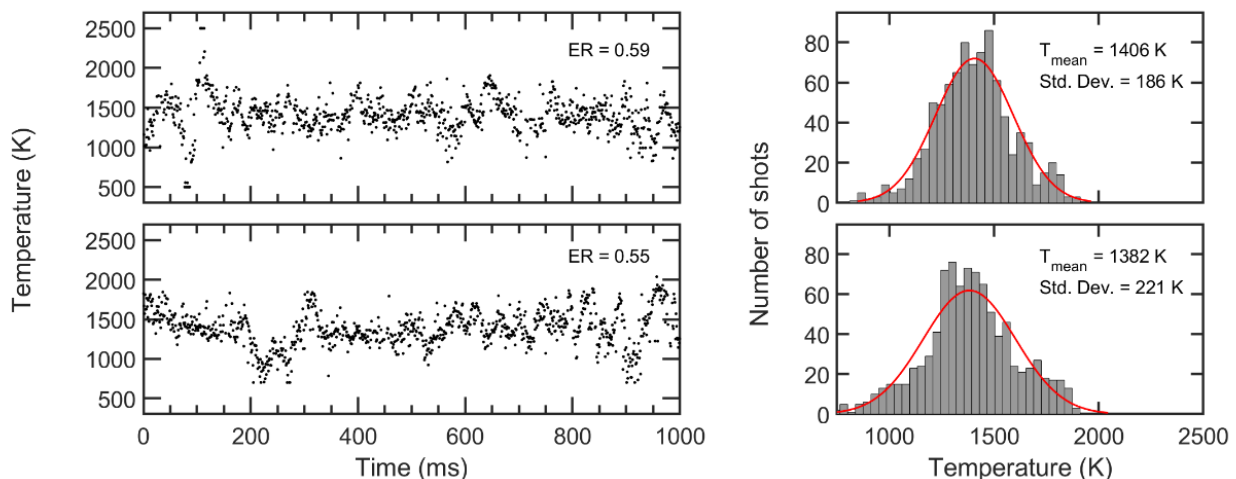
Single-shot CPP fs-CARS spectra were then recorded in the HERON combustion facility for evaluating the CPP fs-CARS performances in high-pressure kerosene/air flames. For this study, a multi-point Low-NO<sub>x</sub> injection system fed with liquid kerosene was used. This one is composed of a single pilot injector located on the combustor axis and it is used to stabilise the flame whereas a multipoint fuel injection is located at larger radial distance. Fuel flowrate is split between the two injectors and the ratio between equivalence ratio from the pilot and the main injector lies between 0.1 and 1, except in the case where only the pilot injector is used. The operating conditions for which CARS measurements were performed are resumed in Table 1.

**Table 1:** Operating conditions

	Air inlet temperature (K)	Pressure (MPa)	Equivalence ratio
Case 1	720	0.75	0.59
Case 2	720	0.75	0.55

Figure 2 shows the time evolution of temperature measurements from 1000 sequential single laser-shots recorded in the region of burnt gases located behind the inner recirculation zone for the two conditions of equivalence ratio. Note in this experiment that the variation in the equivalence ratio

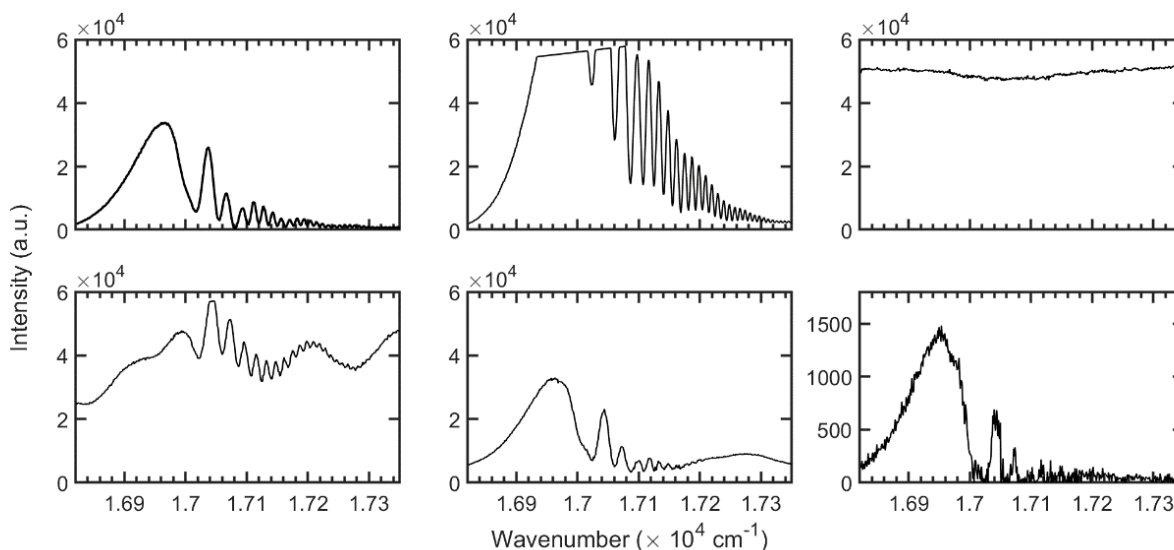
is deliberately small in order to quantify the performance of the CPP-fs CARS in capturing small temperature variations. Each data point denotes a temperature measurement from the data reduction of a single-shot CARS spectrum. The results depicted in Fig. 2 show the time history plots of temperature measurements recorded during 1 sec at the same probe volume. For both cases, the time history profiles show large and fast variations of temperature for 0.01 to 0.1 s in time. These variations, which are easily discernible are the result of intense mixing taking place between the burnt gases produced by the flame and heated inlet air circulating around the periphery of the flame. Origin of this air flow is only related to the operation of the combustion chamber which requires for reasons of thermal resistance, an injection of air along the walls of the combustion chamber. Finally, the right-hand side of Fig. 2 displays the associated probability density function (Pdf) in which the Gaussian envelope is also included. Pdf are built with a temperature column step of 40 K matching the uncertainty of CARS measurements in stationary conditions. A comparison of both Pdfs also shows the capability of the CPP-fs CARS to capture a decrease of the mean temperature when the global equivalence ratio is decreasing while the scale of the rms of temperature fluctuations are of the same order of magnitude.



**Fig. 2:** Time history profiles of 1000 sequential single-shot temperature measurements (left) and associated histograms (right) recorded for two global equivalence ratios.

Depending on the locations in the flame, the  $\text{N}_2$  CARS spectra can present some distortion in shape and in intensity. Fig. 3 illustrates these variations by comparing a  $\text{N}_2$  CARS spectrum without distortion to CARS spectra with different levels of perturbations. Experimental observation indicates that the variation could be related to different reasons such as saturation of the camera, partial interaction with fuel droplets in the CARS probe volume, laser-induced

breakdown in fuel droplets, beam steering effects... In order to minimise these effects, the data reduction method was to examine each CARS spectrum with the genetic algorithm. Once the minimisation routine was used, a discrimination of each CARS spectrum was performed on the basis of the value of the threshold that was tolerated. Above this value, the CARS spectrum was rejected. Such a method, although effective, is unfortunately costly in terms of calculation time. Consequently, the validation rate of CARS measurements, defined as the ratio between the number of spectra successfully processed and the number of laser shots during a run, could be variable according to the locations in the flame.



**Fig. 3:** Examples of CARS signal disturbances inside the combustion chamber.

Two radial temperature profiles were recorded in conditions characteristics of a 0.61 global equivalence ratio and a pressure combustor of 0.75 MPa (Fig. 4). The first profile is recorded at the outlet of the injector in which the flame front is produced while the second one is located in the exhaust gases. For both profiles, the locations of CARS measurements are reported in Fig. 4 by yellow dots. Due to the presence of droplets in the near field of the flame, between 60% and 80% of the single-shot measurements were free from disturbances and data processed. This rate rises to more than 90% in the far field. The histograms of temperature measurements are also shown. In the near field corresponding to the zone where the flame is developing, the scatter in the Pdfs gives insight into the degree of turbulence of combustion that depends primarily on the mixing of kerosene vapour and air inlet. For instance, the probability density functions of temperature within the recirculation zone show a gaussian distribution with a maximum temperature of  $\sim 1150$  K which results from the mixing of air preheated along the wall with combustion products moving in the recirculation zone. When moving towards the flow axis, the mean temperature is gradually



shifted to higher values which can reach  $\sim 1950$  K in the zone of the flame front. We also observe a broadening of the temperature pdfs, these ones showing skewed left or skewed right distributions. Existence of these distributions suggest the presence of gas in the partially burned-unburned state moving randomly in space and time. In the zone of the exhaust gases, the histograms of temperature recorded to the different radial locations become narrower and symmetrical with a temperature peak of about 1700 K. This behaviour simply reflects the degree of homogeneity of the mixing of the exhaust gases produced in the near field and then transported into the recirculation zone.

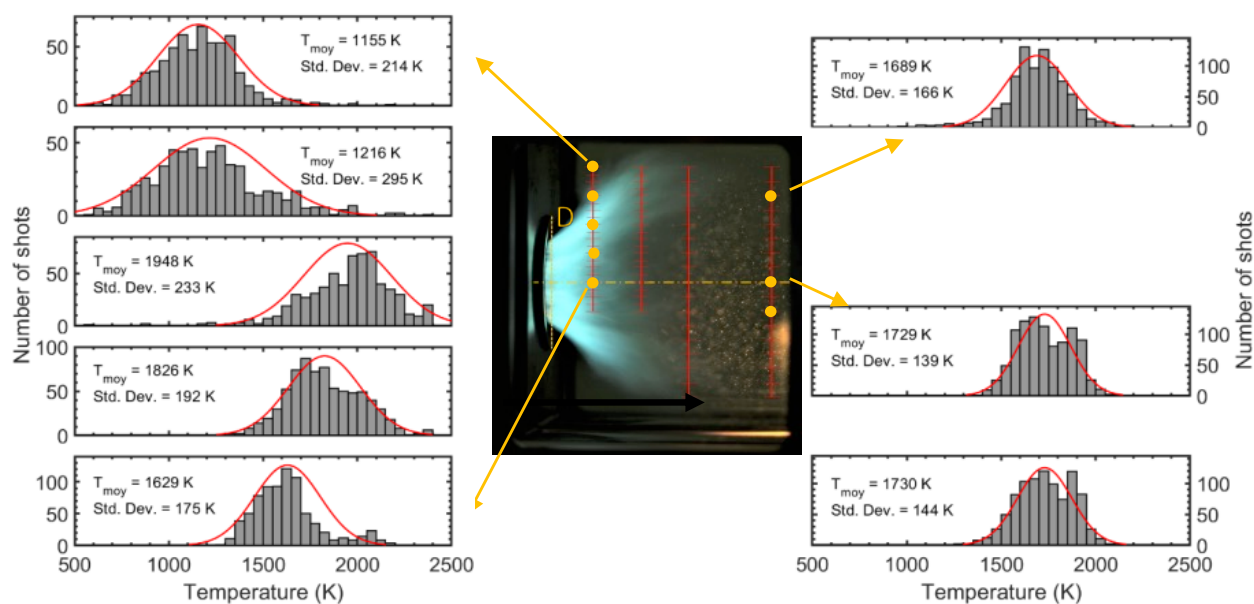


Fig. 4: Temperature distributions in the kerosene/air flame at a pressure condition of 0.75 MPa

## 5. Conclusions

The application of CPP-fs CARS for temperature measurements in a practical model combustor was demonstrated. Measurements were performed at a repetition rate of 1 kHz and the accuracy of the single-shot temperature measurements was about 1.5 % at 2000 K. The mean temperature, the most probable temperature and the temperature fluctuations were used to characterize the performances of a semi-industrial aircraft multi-point injection system fed with liquid kerosene and operating at a pressure of 0.75 MPa. The analysis of the temperature histograms has given valuable information about the homogeneity of the combustion process, the temperature fluctuations about the state of the fuel/air mixing, and the temperature level about the efficiency of the combustion. These results, which are unique to our knowledge, demonstrated the ability of CPP-fs CARS to acquire high-speed temperature measurements in realistic combustors and to

provide benchmark experimental data of high-fidelity for traditional and emerging fuels that will be used to develop and validate the predictive capability of future gas turbine combustors.

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