Dual-pump vibrational CARS measurements of temperature and species concentrations in
turbulent premixed flames with CO₂ addition

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Abstract

A dual-pump vibrational coherent anti-Stokes Raman scattering (DPVCARS) system was applied for simultaneous temperature and species concentration measurements in jet flames on a newly developed piloted axisymmetric reactor assisted turbulent (PARAT) premixed flame burner. Turbulent lean premixed methane/air round jet flames with varying CO₂ dilution were stabilized using an annular flow of hydrogen at the flame base. Carbon dioxide was injected into the fuel stream to simulate effects of dry exhaust gas recirculation (EGR). The flames were operated at a nearly constant Reynolds number, Lewis number, and adiabatic flame temperature to minimize thermal and transport effects. Velocity boundary conditions at the burner exit were measured using particle image velocimetry. Simultaneous temperature and major species concentrations (O₂ and CO₂) were measured along the axial direction at the burner centerline and along the radial direction at representative axial locations above the burner. Progress variables calculated based on the temperatures were used to study the mean flame brush thickness and the turbulent burning velocity. Probability density functions (PDFs) of temperature at various locations are presented and discussed. The DPVCARS measurements provide an experimental

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4 database including temperature and turbulent burning velocity for combustion model evaluation
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6 and validation and demonstrated a strong potential for flame structure investigation with future
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8 improvement on spatial resolution and signal to noise ratio. The merits and limitations of
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10 vibrational CARS for diagnostics in turbulent premixed combustion are discussed. The results
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12 show that flames with CO₂ addition depart from the universal mean flame brush structure, while
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14 their RMS fluctuation values at an identical mean temperature collapse with the undiluted flame.
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16 The CO₂ addition increases the combustion intensity with a decrease in turbulent burning
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18 velocity when minimizing the thermal and transport effects.
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27 **Keywords**

28 Turbulent lean premixed flame; CO₂ addition; Coherent anti-Stokes Raman scattering;
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30 Temperature measurements; Species concentrations measurements
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1. Introduction

Turbulent lean premixed flames are utilized in many applications including stationary gas turbine combustors. The addition of CO₂ into the reactant stream in turbulent premixed combustion process is an important research topic due to its involvement in exhaust gas recirculation (EGR) for pollutant reduction. Tanaka et al. [1] of Mitsubishi Heavy Industries showed that EGR reduces NO_x formation even when the gas temperature at turbine inlet is held constant. The non-thermal effects of diluted gases on pollutant reduction in turbulent premixed combustion have drawn attention from several researchers. The addition of CO₂ to the reactant streams plays an important role in modification of premixed combustion characteristics including flame structure, flame speed, and pollutant formation [2-5]. Le Cong and Dagaut [6] studied the oxidation of methane-based fuels with CO₂ dilution in a jet-stirred reactor for a wide range of equivalence ratios and temperatures. Kinetics of this experiment was modeled using the mechanism proposed earlier by Dagaut [7]. The authors concluded that the consumption of CH₄ was inhibited, mainly, through CO₂ thermal effects whereas chemical effects played a secondary role. The chemical inhibiting effect of CO₂ addition on CH₄ consumption is introduced by perturbation of the equilibrium of the reaction $H + CO_2 = CO + OH$ [6]. By studying laminar spherical flames with CO₂ addition, Xie et al. [8] also pointed out that CO₂ directly participates in chemical reactions through the elementary reaction $H + CO_2 = OH + CO$ and inhibits the combustion process because of the competition for the H radical between the reaction $H + CO_2 = OH + CO$ and the reaction $H + O_2 = O + OH$. The effects of CO₂ addition on laminar flame speed were examined by Zhu et al. [3]. They reported a decrease in the laminar flame speed with CO₂ addition at identical adiabatic flame temperatures. Extinction characteristics of CO₂ diluted premixed flames were studied by Ju et al. [9] and Chen et al. [10]. They found that the reabsorption of emitted radiation due to the presence of CO₂ in fresh gases leads to higher

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4 burning velocities and wider extinction limits. Fackler et al. [5] suggested that premixed flames
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6 diluted with CO₂ produce less NO_x due to both a decrease in N₂ concentration and the depletion
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8 by CO₂ of radicals participating in NO_x formation chemistry.
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11 In addition to these studies of CO₂ dilution effects in laminar premixed flames, a few studies
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13 of these effects in turbulent premixed flames have also been reported. Kobayashi et al. [11] used
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15 OH PLIF to study CH₄-CO₂-air flames with the mixture preheated up to 573 K at operating
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17 pressures up to 1 MPa. They concluded that the combustion intensity and the mean fuel
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19 consumption rate decreased with an increase in CO₂ addition. Using Mie scattering tomography,
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21 Cohe et al. [12] reported that the flame wrinkling parameters and flame surface density did not
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23 change significantly with an increase in CO₂ dilution. However, inconsistent with the results in
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25 reference [11], they observed an increase of combustion intensity with CO₂ addition.
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27 Simultaneous quantitative measurements of temperature and species distributions and their
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29 statistical properties in turbulent premixed flames with CO₂ addition have not been reported in
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31 the literature.
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39 Measurements of the statistics of temperature distributions are important in turbulent
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41 premixed flames because of their impact on chemical reaction rates, instabilities, and pollutant
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43 formation. The measurements allow experimental validation of combustion models, especially
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45 large eddy simulation (LES) models [13]. Coherent anti-Stokes Raman scattering (CARS) has
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47 been proven to be a useful technique for point-wise temperature measurements in gas-phase
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49 reacting flows [14, 15]. In the past, several investigations have been conducted using N₂
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51 vibrational CARS (VCARS) thermometry in turbulent flames [16-18]. For example, Magre et al.
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53 [18] obtained temperature statistics for both premixed flames and hot non-reacting jets. A key
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55 issue is that the dimension of the probe volume along the propagation direction of the laser
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beams is, typically, larger than the instantaneous flame characteristic length, where the temperature changes rapidly from that of reactants to that of products. In regions of such high-temperature gradients, the CARS signal may have contributions from gases at different temperatures. This spatial averaging effect has been investigated under various conditions [19-23]. A quantitative method to correct the temperature measurements under spatial averaging effects in turbulent flames has not been developed. This is because such a correction requires the knowledge of the unsteady gas composition within the probe volume. It is noted that the overlap and focusing of multiple laser beams allow CARS better spatial resolution than other measurement systems. Details of the spatial resolution of the CARS system used in this study and its impact on the measurements are discussed in a subsequent section.

Concentrations of the major gas species O_2 and CO_2 were measured simultaneously with temperature by dual-pump vibrational coherent anti-Stokes Raman scattering (DPVCARS). These measurements can provide critical information for flame characterization as well as for combustion modeling. Hancock et al. [24] measured temperature and O_2 concentration in a laminar jet flame using CARS. Frederickson et al. [25] measured temperature and O_2 concentrations in a turbulent pool fire. Both groups found that the CARSFT model overestimated O_2 concentrations at high temperatures. This was attributed to inaccurate modeling of Raman linewidths for higher-J Q branch rotational lines. Magnotti et al. [26] probed N_2 , O_2 , and H_2 in a supersonic combustion system. Good agreement between measured O_2 concentration and theory was shown in their calibration measurements on a Hencken burner. Carbon dioxide concentrations and temperatures have also been probed in counterflow diffusion flames [27], CO_2 seeded H_2 diffusion flames [28], exhaust streams of a liquid-fueled combustor [29], flat flames [30, 31], and in a gas cell [30]. Both higher and lower measured CO_2

concentrations than those from calculations were observed. In addition to the effects of approximate line-width models, the accuracy of the species concentration measurements in turbulent flames is limited by spatial averaging effects.

In this study, a dual-pump vibrational CARS (DPVCARS) system was used for simultaneous temperature and species concentration (O_2 and CO_2) measurements in turbulent premixed flames stabilized on a newly developed piloted axisymmetric reactor assisted turbulent (PARAT) burner. Hydrogen-piloted turbulent lean premixed methane/air flames with CO_2 addition were investigated. Carbon dioxide was injected into the reactant stream to simulate dry EGR. The adiabatic flame temperature, Reynolds number, and Lewis number of the flames investigated were constant within estimated uncertainties. Single-shot temperature and species concentration measurements were obtained using DPVCARS. The inflow velocities were measured using a particle image velocimetry (PIV) system [32] in conjunction with software for robust phase correlation [33]. The objective of this work is to use DPVCARS and PIV measurements to advance our fundamental understanding of the structure of turbulent premixed flames with CO_2 addition. The merits and limitations of applying DPVCARS for studies of turbulent premixed flames are discussed. A database of measurements has been generated for future development and validation of combustion models for computations including LES [34, 35] for gas turbine combustion with dry EGR.

2. Experimental methods

2.1. Burner details and operating conditions

A newly designed piloted axisymmetric reactor assisted turbulent (PARAT) burner [36] was used to stabilize lean, turbulent, CH_4 /air premixed flames with varying amounts of CO_2 dilution. The burner can be operated at both atmospheric and high-pressure conditions up to 20 bars. A

schematic diagram of the cross-section of the PARAT burner is shown in Fig. 1. The nozzle exit has an 18 mm diameter (D) annular cross section with a 3-mm thick wall. The burner is 523 mm tall with diverging (bottom) and converging (near the nozzle exit) sections. The diverging and converging nozzles were designed to prevent flow separation and recirculation zones while also reducing the flow boundary layer thickness. To sustain a flame at large Reynolds numbers, a non-premixed pilot flame of undiluted hydrogen was utilized. The pilot flame was generated by flowing hydrogen through a 2 mm wide annulus just outside of the nozzle outer wall.

Turbulence generator plates were installed within the burner [36]. Schematic diagrams of the turbulence generator plates are shown in Fig. 2. The lower turbulence generator plate is located before the final convergence of the flow channel, 177 mm upstream of the nozzle exit. This plate has 5 circular holes with a diameter of 6.35 mm and a 94% blockage ratio. The upper turbulence generator plate is placed 30 mm upstream of the nozzle exit. The upper plate has 13 circular holes with a diameter of 2.54 mm and an 87% blockage ratio. This turbulence generator with smaller holes governs the “integral” scale of turbulence and serves as a flame holder in the event of a flame flashback.

The burner was translated in the vertical direction using a manual lab jack and in the horizontal directions using a computer-controlled positioner (Zaber A-LST0250A) for measurements at different locations within the flame. The cameras and optics were stationary. The burner includes an air inlet at the bottom and fuel and CO₂ inlets further upstream. A thermocouple was installed in the contraction nozzle to detect any accidental flashback.

Three atmospheric pressure conditions were chosen to study flame structure with and without CO₂ addition as shown in Table 1. The laminar flame speeds were calculated using CHEMKIN with the GRI 3.0 mechanism. The laminar flame thermal thicknesses (characteristic flame

thickness) for the flames were estimated, following Lipatnikov [37], within the framework of the activation energy asymptotic theory. The flames were operated at identical jet Reynolds numbers (10,000) and the equivalence ratios were chosen to yield identical adiabatic flame temperature (2030K) in order to minimize the thermal effects on flame propagation and structure. The peak mean temperatures for all three flames were measured to be identical within experimental error. The unburned mixture Lewis numbers differed by a maximum of 2%, thereby minimizing transport effects on the flame structure. Therefore, any differences in the flame structure and the temperature distributions are expected to be due to the effects of CO₂ addition. The CH₄, CO₂, and H₂ flow rates were controlled by digital mass flow controllers with a precision of about 2%. The air flow rate was determined by the upstream pressure gauge of an orifice plate which was calibrated using a dry test turbine meter. The heat release rates of the pilot H₂ flame and CH₄ flame were calculated based on their flow rates and lower heating values. The nominal jet exit Reynolds numbers are based on the mixed cold gas properties [38], the exit velocity of the central jet burner, and the burner diameter. Velocity boundary conditions and turbulence statistics were measured using a digital PIV system. Temperatures near the tip of flames 2 and 3 were measured twice for evaluating the repeatability of the CARS data.

2.2.Coherent anti-Stokes Raman scattering system

A dual-pump vibrational coherent anti-Stokes Raman scattering (DPVCARS) system developed by Satija and Lucht [39] was used to simultaneously measure temperature and O₂ and CO₂ concentrations. This system employed 532 nm and 561 nm beams as interchangeable pump/probe beams and a broadband dye laser at 607 nm as the common Stokes beam. The 532 nm beam was obtained by frequency doubling the fundamental beam from Nd: YAG laser. The 561 nm beam was generated using a commercial narrowband dye laser whereas the 607 nm beam was generated using a broadband dye laser with a bandwidth of approximately 50 nm. The

DPVCARS signal was generated at 496 nm. The energy of the input beams was adjusted according to the measurement location in order to avoid saturation and Stark effects [40] while acquiring signals with sufficient signal to noise ratio (SNR). The 532 nm, 561 nm, and 607 nm beams contained approximately 20 mJ/pulse, 20 mJ/pulse, and 30 mJ/pulse of energy respectively, and the diameter of all the beams, prior to the focusing lens defining the probe volume, was approximately 8 mm. The CARS system had a spatial resolution of 150 μm x 1.5 mm and a temporal resolution of 6 ns. The spatial resolution is defined as the FWHM of the non-resonant signal obtained by scanning a very thin coverslip along the direction of the propagation of the laser beams. At each spatial location in the flame, 1000 single shots were collected to generate sufficient data for studying temperature statistics and correlations between species concentrations and temperature.

2.3. Analysis of CARS experimental data

Temperature was determined as a fitting parameter in a least squares fitting of calculated to experimental single shot CARS spectra using the CARSFT [41, 42] code. The single-shot precision of the temperature measurement was assessed in laminar flames on a Hencken burner [43]. The single-shot precision was 2% near 2000K, about 5% near 1000 K, and about 8% at room temperature. The single-shot O₂ spectra were fit using the model employed by Hancock et al. [24]. That model has been found to overestimate O₂ concentration at higher temperatures [24]. This overestimation was attributed to inaccurate modeling of Raman linewidths for higher-J Q branch rotational lines. The CO₂ spectra were fit using the CARS model employed by Lucht et al. [30]. The accuracy of this model has been assessed, in Hencken burner flames, to be about 10%.

Two representative experimental single shot spectra recorded at $r/D=0$ and $x/D=5.17$ above the PARAT burner exit are shown in Fig. 3. The parameter r is the distance from nozzle centerline and the parameter x is the distance from the nozzle exit plane. The lower-temperature spectrum shows clearly distinguishable CO_2 , N_2 , and O_2 peaks. Excellent fits of the single-shot nitrogen spectra to the theoretical spectra were observed. In the high-temperature spectrum, both fundamental and hot bands of N_2 are visible. However, at high temperatures (>1800 K), O_2 and CO_2 lines were difficult to quantify due to a low signal to noise ratio. Besides the low signal to noise ratio, the spatial averaging effect also influences the quality of the fit. Good quality fits were always observed for N_2 spectra. However, O_2 and CO_2 spectra did not always fit well due to spatial averaging effects and due to poor SNR at higher temperatures. Therefore, PDFs of O_2 and CO_2 were not constructed. Single-shot based correlations between O_2/CO_2 concentrations and temperatures are plotted for spectra where acceptable fits and compared with corresponding values from flame theory.

A quantitative analysis of the spatial averaging effect is out of the scope of this work. Qualitatively, however, we do not expect a strong influence of spatial averaging on measurements outside the flame brush. Hence, the measured peak flame temperatures for all the flames are expected to be accurate to within 2%. Inside the flame brush, due to stronger temperature and concentration gradients, the measured temperatures have a larger uncertainty. The presence of intermediate temperatures within the temperature PDFs at locations inside the flame brush can be partly attributed to both finite flame thickness and the spatial averaging effects.

2.4. Particle image velocimetry system

A particle image velocimetry (PIV) system [32] was employed to characterize the inflow boundary conditions. This system consists of a double-pulsed, frequency-doubled, Q-switched Nd:YAG laser (Spectra-Physics Quanta-Ray GCR200 PIV400-10), an optical system, and a high-speed CCD camera (PIVCAM 10-30, TSI Inc.) with a 300mm/F3.5 lens (Tamron AF 28-300 mm F3.5). The laser operates at 532 nm and emits 400 mJ per pulse at 10Hz, with a pulse duration of 9 ns. A laser-pulse synchronizer (Model: 610034, TSI Inc.) was used to synchronize the laser and the CCD camera. The double-pulsed laser beams were formed into a sheet using a combination of spherical and cylindrical lenses. The light scattered by the tracer particles was recorded by the high-speed CCD camera with a resolution of 1000 x 1016 pixels at 10 frames/s. The laser pulse interval was set to 12 μ s. The imaging region was set to 26 mm $\times$ 26 mm, with each pixel corresponding to 26 μ m. The laser sheet thickness was about 300 μ m based on a photographic paper burn pattern. The central jet flow was seeded with alumina particles with a nominal diameter of 0.5 μ m. One thousand image pairs were captured for statistical analysis.

The 2D vector fields from the PIV images were evaluated using the PRANA tool with Robust Phase Correlation [33]. An adaptive window offset with iterative image deformation algorithm was employed to minimize noise and to allow more resolved measurements. The final interrogation window was 20 $\times$ 20 pixels with 75% overlap, resulting in a spatial resolution of 0.52 mm and a vector spacing of 0.13 mm. A three-point Gaussian-fitting algorithm was used to provide sub-pixel accuracy for the determined velocity. Less than 3% of vectors were found as outliers (erroneous vectors) in the final pass using universal outlier detection method. Most of the outliers were located inside the shear layer at the edge of the jet due to lack of seeding particles. In post-processing, vectors were compared to the mean of all adjacent vectors. Vectors more than 3 standard deviations from the mean were removed from the ensemble for

mean and RMS calculation. Interpolated vectors to place outliers were only used for integral length scale calculation. Standard uncertainties of the measurements were estimated based on the cross-correlation peak ratio from single shot measurements [44]. The uncertainty for the velocity magnitude was decomposed into axial and radial direction based on the local velocity to provide a separate assessment axial and radial velocity measurement. The PIV measurements do not resolve the smallest scale of velocity fluctuations (~ 0.05 mm). Therefore, the uncertainties in the turbulent fluctuation measurements result from both spatial averaging effects and measurement uncertainties.

3. Results and discussion

3.1 Velocity boundary conditions and turbulence statistics

The measured velocity profiles at $x/D=0.2$ are used to characterize inflow boundary conditions and turbulent statistics as shown in Fig. 4. The mean axial velocity profiles are within uncertainty limits for all three flames with identical Reynolds number. Representative error bars showing standard uncertainty are shown for only every tenth point for clarity. The RMS axial velocity fluctuation values are nearly identical for the three flames. A constant value of about 2 m/s of RMS axial velocity fluctuation is observed within the burner diameter. The mean radial velocity profiles show a similar behavior for all three flames. The absolute values of the mean radial velocities are an order of magnitude smaller than those of the mean axial velocities. However, the RMS radial fluctuation values, which are not plotted in Fig. 4, are of similar magnitude at about 1.6 m/s within the burner diameter for all three flames. These are best expressed in terms of the total turbulence fluctuation values and turbulence intensity defined as

$$u_{rms} = \sqrt{\frac{u_{x,rms}^2 + 2u_{r,rms}^2}{3}} \text{ and } T.I. = \frac{u_{rms}}{u_{mean}} \text{ for an axisymmetric flow field, where } u_x \text{ and } u_r \text{ are the}$$

axial and radial velocity components. The RMS fluctuation values of the velocity are slightly

lower than the RMS fluctuation values of its axial component due to smaller RMS fluctuation values of the radial and azimuthal components. The measured turbulence intensities values are nearly constant of about 14 - 15% between shear layers for flames 1, 2, and 3 at $x/D=0.2$. Inside the shear layers, the turbulence intensity increases due to the decrease in local mean velocity. The integral length scale is calculated based on two-point correlation of u_x component of velocity

as $l(r) = \int_0^\infty \rho(r, r^*) dr^*$, where the two-point correlation is defined as:

$$\rho(r, \Delta r) = \frac{\overline{u'_x(r)u'_x(r+\Delta r)}}{\overline{u'^2_x(r)}}; \Delta r = |r - r^*|. \text{ Numerical integrations of the measured velocities at a}$$

point and at points with incrementally larger radii are completed to obtain statistically significant values of the length scale and the autocorrelation coefficient. The measured integral scales at $x/D=0.2$ are about 1 mm for flames 1, 2, and 3. The integral length scales estimated from the two-point correlation coefficients depict approximately 20% noise based on compounding of noise at multiple points. However, the estimated integral length scales are still of value in support of premixed combustion model developments. The integral length scales are comparable to the diameter of the holes on the upper turbulence plate.

3.2 Temperature

The measured values of the mean and RMS temperature as a function of the distance along the centerline axis are plotted in Fig. 5. The mean temperature along the axis remains constant (within 5 K) and equal to the reactant temperature for the initial ~1.4 diameters for the flames with 0%, 5%, and 10% CO_2 addition. The temperature remains almost constant due to the lack of diffusion and chemical reactions within the potential core region of the jet. The lengths of the potential cores of the three flames are identical within uncertainty because of the identical Reynolds number and mixture properties. The RMS of the temperature distribution also remains

constant at around 80 K. The low RMS of temperature fluctuations observed inside the unburned reactant region is due to the lack of mixing between the reactants and the products. Downstream of this region, the mean temperatures of all three flames increase. The RMS temperature fluctuations initially increase with axial distance to a maximum and then decrease to a minimum as the location of the maximum mean temperature is approached. The variations in RMS fluctuations result from the evolution in probabilities of finding reactants and products. In the post-flame region, the RMS temperature fluctuations increase with decreasing mean temperature because of the buoyant mixing of the combustion products and the ambient air. The mean flame length (defined as the distance from burner exit to the location of the maximum mean temperature) was found to increase with an increase in CO₂ dilution.

The mean and RMS temperatures for flames 1, 2, and 3, as a function of the radial distance (r/D) at axial locations of $x/D=1.94$ and 5, are shown in Fig. 6. At the axial location of $x/D=1.94$, which is closer to the burner exit, a cold core and an annular region consisting of an increase to the flame temperature peak and then a decrease as a result of mixing with the surrounding air are observed in the mean temperature plots for both flames. Lower mean temperatures are observed for the flame with 10% CO₂ addition at identical radial locations. At $x/D=5$, the mean temperature profile of the flame near the center, with 0% CO₂ addition, shows a temperature close to that of the completely burned products. The temperature decreases along the radial direction indicative of a mixing process between the combustion products and ambient air. However, the mean temperature profile of the flame with 10% CO₂ addition has a local minimum near the axis which indicates the presence of unburned reactants and a longer flame length along the axial direction.

The mean temperatures were used to calculate the mean progress variable, defined as

$$C_{mean} = \frac{T_{mean} - T_{mean,min}}{T_{mean,max} - T_{mean,min}} .$$

The mean progress variable is plotted as a function of a non-dimensional distance ξ , defined in the following, in Fig. 7. The mean flame brush thickness, δ_i , was determined using the maximum gradient method [37]. The formulae for δ_i are

$$\delta_i = \max^{-1} \left(\frac{dC_{mean}}{dx} \right) \text{ for the centerline and } \delta_i = \max^{-1} \left(\frac{dC_{mean}}{dr} \right) \text{ for the radial direction.}$$

The maximum gradient was calculated at the data points closest to $C_{mean} = 0.5$. The error caused by a limited number of measurements, especially for the radial measurements where fewer data points were collected, is expected to be about 10%. An analysis of the mean flame brush thickness is not only important for numerical model validation but is also meaningful for turbulent combustion regime characterization. In the literature, several measured spatial profiles of progress variable, presented as a function of the dimensionless variable ξ , show a collapse to a universal curve for a wide range of conditions [45]. The dimensionless distances ξ are defined

$$\text{as } \xi = \frac{x(C_{mean}) - x(C_{mean} = 0.5)}{\delta_i} \text{ along the centerline and } \xi = \frac{r(C_{mean}) - r(C_{mean} = 0.5)}{\delta_i} \text{ along the}$$

radial direction. A solid curve calculated using the complementary error function parameterization as $C_{mean} = 0.5 \operatorname{erfc}(-\xi \sqrt{\pi})$ is also plotted for comparison as shown in Fig. 7.

The parameters used for calculating the dimensionless distance ξ are shown in Table 2.

Lipatnikov [46] examined the universality of mean flame brush structure using the complementary error function expression for C_{mean} above for several undiluted turbulent

premixed flames. Another parameterized form $C_{mean} = \frac{1}{1 + \exp(-4\xi)}$ has also been used by other

researchers [12, 47]. The two curves are very close to each other for the present conditions and therefore the complementary error function is shown in Fig. 7. In Fig. 7, the parameterized function matches well for the flame without CO₂ addition, Flame 1. However, deviations are observed for flames with 5% and 10% CO₂ additions. The solid curve matches the experimental data at lower values of the mean progress variable ($C_{mean} < 0.6$) but the data points fall below the solid curve for $C_{mean} > 0.6$. For $C_{mean} > 0.6$, the mean progress variable profiles of the flames with different levels of CO₂ addition match with each other. Fig. 7 also shows self-similarity of C_{mean} profiles [46]. The spatial profiles of the mean progress variable along the centerline and along the radial direction are similar.

The turbulent burning velocity (or global consumption speed [48]) is another important characteristic of premixed turbulent combustion. The turbulent burning velocity was estimated using the mean progress variable profiles along the burner centerline derived from temperature measurements. The evaluation of turbulent burning velocity is sensitive to the selection of reference surface defined as a surface of constant C_{mean} [48]. The angle method [12, 49] was used for evaluating the turbulent burning velocity. The turbulent burning velocity is written as:

$$S_T = \sqrt{\frac{U^2}{1 + [2x(C_{mean} = 0.2) / D]^2}}, \text{ where } U \text{ is the bulk velocity of unburned reactants. The}$$

presumed cone shape of the C_{mean} contour requires the reference surface to be closer to the reactant side in order to reduce the error caused by the roundish shape of constant C_{mean} contour as shown in references [12, 49]. On the other hand, the reference C_{mean} value needs to be selected with small curvature on the spatial profile to minimize the interpolation errors. Therefore, $C_{mean}=0.2$ was chosen to determine the turbulent burning velocity as an example (similar trends were obtained by using $C_{mean}=0.3, 0.4$, and 0.5). Figure 7 shows the turbulent

burning velocity and combustion intensity (defined as turbulent burning velocity normalized by unstretched laminar flame speed, S_L [12]) as a function of u_{rms} / S_L . A parameterized function [45] is plotted to fit the measured combustion intensities. The turbulent burning velocity decreases with CO₂ addition while the combustion intensity increases with CO₂ addition. The latter observation agrees with the measurements of Cohe et al. [12]. A nonlinear dependence of S_T on S_L is shown in the bending region of the fitted curve. Shy et al. [50] reported that the bending effect promoted by CO₂ addition in rich turbulent premixed flames is due to the radiative heat loss.

RMS temperature fluctuation values are plotted as a function of the mean temperature in Fig. 9. The RMS of a purely wrinkled flame (zero instantaneous flame thickness) is shown as a curve for comparison. The RMS for a purely wrinkled flame can be expressed as $T_{RMS} = \sqrt{(T_{max} - T_{mean})(T_{mean} - T_{min})}$, where T_{max} is the adiabatic flame temperature, and T_{min} is the unburned reactant temperature [18]. Similar methods were employed by Driscoll and Gulati [51] who used RMS density fluctuation values and Chen and Bilger [52] who used RMS progress variable fluctuation values to compare measurements with the thin flame limits. The RMS temperature fluctuation values for all three flames show similar deviation from the value estimated from the purely wrinkled flame theory. The deviation remains consistent when moving either along the axial or the radial direction. The length of the CARS probe volume being greater than the instantaneous flame thickness contributes to a part of the deviation from the thin flame limit. The RMS temperature fluctuation values, as shown in Fig. 9, are probably less than the true value within the flame brush region. The deviations of the RMS profile result from both spatial averaging effects and the finite thickness of the instantaneous flames.

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4 However, the influence of a larger laminar flame thermal thickness resulting from CO₂ addition,
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6 as shown in Table 1, is not evident from Fig. 9.
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10 Probability density functions (PDFs) of temperature from flame 1, flame 2, and flame 3 at
11 different axial locations along the centerline are presented in Fig. 10. These PDFs show an
12 evolution of temperature statistics from those for the unburned reactants to those within the
13 flame brush, those for completely burned products, and finally for the cooled and diluted
14 products when mixed with ambient air. The shape of the PDF for flame 1 in Fig. 10 in the
15 bottom panel indicates a large probability of finding unburned reactants. As the flow moves
16 further downstream to the flame brush ($x/D=3.11$), the shape of the PDF switches to a bimodal
17 distribution. This distribution indicates a significant probability of finding both unburned
18 reactants and burned products due to the fluctuations in the location of the reaction zone.
19 Furthermore, a finite probability of intermediate temperatures is found in this distribution
20 otherwise dominated by the bimodal values of reactant and product temperatures. These
21 intermediate temperatures are most likely to result from the measurements of both cold and hot
22 gases inside the probe volume considering the instantaneous flame thickness to be less than the
23 probe volume size. At $x/D=3.67$, the PDF presents a Gaussian-like distribution centered at the
24 mean burned product temperature with finite probabilities of low and intermediate temperatures.
25 Closest to the location ($x/D=5.17$) of the completely burned products, the shape of the PDF
26 shows a clipped Gaussian- distribution centered at the mean adiabatic product temperature. The
27 width of the PDF is due to both turbulent combustion as well as the single-shot precision of the
28 CARS system. Beyond the flame tip at $x/D=7.39$, the PDF of temperature shows a high
29 probability of intermediate temperatures mainly resulting from the mixing of the combustion
30 products with the ambient air. The general trends observed for flame 1 are also observed for
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4 flame 2 and flame 3. However, the evolution of the PDFs is spatially delayed due to the
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6 presence of CO₂ in the fuel stream.
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10 The PDFs of temperature for flame 1 and flame 3 at four representative radial locations are
11 shown in Fig. 11. The progression of the shapes of the PDFs in the radial direction is similar to
12 their progression in the axial direction. The PDFs for flame 1 and flame 3 at identical radial
13
14 locations show some differences. At $r/D=0.28$, the PDF for flame 1 shows that there is a finite
15 probability of finding products while the PDF for flame 3 shows mostly unburned reactants. At
16
17 $r/D=0.5$, the PDF of flame 1 resembles a Gaussian distribution centered at the mean product
18 temperature but that of flame 3 is bimodal. The spatial delay in flame development of flame 3
19 due to CO₂ addition leads to this behavior. At $r/D=0.72$, PDFs for both flame 1 and flame 3
20 show broadened clipped Gaussian distributions with a shorter tail, compared to that at $r/D=0.5$,
21 on the reactant temperature side. At $r/D=0.94$, the PDF of temperatures spreads between
22 ambient and ~2100 K for flame 1 while that for flame 3 spreads between ambient and ~1800K.
23
24 This behavior results from a faster cooling process of combustion products by the ambient air for
25 flame 3 compared to flame 1. The radial flux of products generated by flame 3 (with slower
26 local burning velocity) is lower than that generated by flame 1 resulting in the PDFs described
27 above.
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46 **3.3 CO₂ and O₂ concentrations**

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48 The relationships between measured temperatures and CO₂ and O₂ concentrations are
49 examined in Fig. 12 and Fig. 13 respectively. CO₂ and O₂ concentrations, calculated using the
50
51 Bray-Moss-Libby (BML) model [53] and assuming an adiabatic mixing process are compared
52 with the experimental data. Figure 11 shows three representative panels inside the flame brush,
53 near the peak mean temperature, and in the post flame mixing region. Most CO₂ data at the
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4 highest temperatures ($>1800\text{K}$) are not plotted because of low SNR. At $x/D=3.11$, the CO_2
5
6 measurements at intermediate temperature are mostly filtered out according to the poor fit
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8 quality criteria due to the spatial averaging effects. The spatial averaging effects as observed in
9
10 the spectra are especially pronounced for CO_2 due to its strong 0000-1000 transition
11
12 (overlapping Q-branch lines in transition). Therefore, at $x/D=3.11$, only low-temperature data
13
14 are shown. At $x/D=5.17$ only 13 data points satisfied the fit quality criteria because the
15
16 temperature is above 1800 K for the rest of the data points. At $x/D=7.39$, many more scatter
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18 points are observed compared with the data from the lower axial positions discussed above.
19
20 Because half of the temperature data were under 1800 K and these measurements are located in
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22 the region where the intermediate temperatures mostly result from the mixing between hot
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24 products and ambient air. The measurements in the post flame region agree with the theoretical
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26 values to within 10%, which is approximately the precision of the measurements based on the
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28 literature [30, 54].
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36 Figure 12 shows the correlation between the measurements of temperature and O_2
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38 concentration. The same screening methods applied to CO_2 data were also used for the O_2
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40 measurements. At $x/D=3.11$, O_2 concentrations are shown mostly at low and intermediate
41
42 temperatures. The O_2 concentrations at lower temperature show a good match with the
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44 CHEMKIN calculations. At this location, more O_2 data points were obtained in comparison with
45
46 the CO_2 data points because of the lower sensitivity of O_2 to the spatial averaging effects. At
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48 $x/D=5.17$, very few O_2 concentration data points could be shown because the concentrations are
49
50 very low in most pockets with high temperatures ($>1800\text{ K}$). At $x/D=7.39$, good fits were
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52 obtained to the spectra with minimal spatial averaging effects, therefore, most of the data points
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54 under 1800 K are plotted. With increasing temperatures increasing discrepancies between the
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experimental results and the CHEMKIN calculations are observed. This is probably due to the inaccuracy of the CARS O₂ model because of the uncertainty in the Raman linewidths for high-J Q-branch transitions. The data for O₂ concentrations in flames 2 and 3 were found to be very similar, overall, to those for flame 1.

4. Summary and Conclusions

A DPVCARS system was used to measure single-shot temperatures and species concentrations in piloted turbulent lean premixed methane/air flames with CO₂ addition. The potential for using DPVCARS for diagnostics in turbulent flame combustion is shown and important issues such as spatial averaging are discussed. A database including temperature and turbulent burning velocity has been generated for combustion model evaluation and validation including the effects of dry EGR. The flames were operated with almost identical Reynolds numbers and adiabatic flame temperatures to minimize thermal and transport effects. PIV was employed to provide inflow velocity boundary conditions. Mean and RMS temperature profiles were plotted along the centerline and radial direction at representative axial locations. A progress variable calculated based on temperature measurements was used to evaluate the mean flame brush thickness and the turbulent burning velocity. Single-shot CARS data were used to generate PDFs of temperature and to extract correlations between temperature and species concentrations.

From the temperature data, an increase in the mean flame length was observed with CO₂ addition. Reductions in the mean flame brush thickness, with CO₂ addition, were found from radial temperature profiles. The spatial profile of progress variable shows good agreement with the universal profile established in the literature for flame 1 with no CO₂ dilution but deviations were observed for flames with CO₂ addition. Self-similarity of mean flame brush profile was

observed for flames with and without CO₂ dilution. The turbulent burning velocity decreased with CO₂ addition, while an increase in combustion intensity was observed. The RMS temperature fluctuation values as a function of mean temperature show similar profiles for all three flames. An assessment of the deviation from a purely wrinkled flame theory was affected by the spatial averaging effects, which make it difficult to assess the finite thickness of the instantaneous flame. The PDFs of temperature showed a similar evolution for the flames with and without CO₂ addition. A bimodal distribution was observed within the flame brush. The correlations between the measured O₂ mole fractions and the measured temperatures show a deviation of 10% over the calculations most likely because of the over-prediction of the O₂ concentrations by the CARSFT code. The correlation between measured CO₂-concentrations and temperatures, while less clear at pre-flame locations due to the lack of data points, show a good agreement with the Bray-Moss-Libby (BML) (53) model based calculations in the post-flame region.

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Reference

- [1] Y. Tanaka, M. Nose, M. Nakao, K. Saitoh, E. Ito, K. Tsukagoshi, Development of low NO_x combustion system with EGR for 1700 C-class gas turbine, Mitsubishi Heavy Industries Technical Review 50 (2013) 1.
- [2] T. Le Cong, P. Dagaut, Oxidation of H₂/CO₂ mixtures and effect of hydrogen initial concentration on the combustion of CH₄ and CH₄/CO₂ mixtures: Experiments and modeling, Proceedings of the Combustion Institute 32 (2009) 427-435.
- [3] D.L. Zhu, F.N. Egolfopoulos, C.K. Law, Experimental and numerical determination of laminar flame speeds of methane/(Ar, N₂, CO₂)-air mixtures as function of stoichiometry,

pressure, and flame temperature, Symposium (International) on Combustion 22 (1989) 1537-1545.

[4] Y. Ju, H. Guo, K. Maruta, F. Liu, On the extinction limit and flammability limit of non-adiabatic stretched methane–air premixed flames, *Journal of fluid mechanics* 342 (1997) 315-334.

[5] K.B. Fackler, M.F. Karalus, I.V. Novosselov, J.C. Kramlich, P.C. Malte, Experimental and numerical study of NO_x formation from the lean premixed combustion of CH₄ mixed with CO₂ and N₂, *Journal of Engineering for Gas Turbines and Power* 133 (2011) 121502-121502-121507.

[6] T.L. Cong, P. Dagaut, Experimental and detailed kinetic modeling of the oxidation of methane and methane/syngas mixtures and effect of carbon dioxide addition, *Combustion Science and Technology* 180 (2008) 2046-2091.

[7] P. Dagaut, On the kinetics of hydrocarbons oxidation from natural gas to kerosene and diesel fuel, *Physical Chemistry Chemical Physics* 4 (2002) 2079-2094.

[8] Y. Xie, J. Wang, M. Zhang, J. Gong, W. Jin, Z. Huang, Experimental and numerical study on laminar flame characteristics of methane oxy-fuel mixtures highly diluted with CO₂, *Energy & Fuels* 27 (2013) 6231-6237.

[9] Y. Ju, G. Masuya, P.D. Ronney, Effects of radiative emission and absorption on the propagation and extinction of premixed gas flames, Symposium (International) on Combustion 27 (1998) 2619-2626.

[10] Z. Chen, X. Qin, B. Xu, Y. Ju, F. Liu, Studies of radiation absorption on flame speed and flammability limit of CO₂ diluted methane flames at elevated pressures, *Proceedings of the Combustion Institute* 31 (2007) 2693-2700.

[11] H. Kobayashi, H. Hagiwara, H. Kaneko, Y. Ogami, Effects of CO₂ dilution on turbulent premixed flames at high pressure and high temperature, *Proceedings of the Combustion Institute* 31 (2007) 1451-1458.

[12] C. Cohé, C. Chauveau, I. Gökalp, D.F. Kurtuluş, CO₂ addition and pressure effects on laminar and turbulent lean premixed CH₄ air flames, *Proceedings of the Combustion Institute* 32 (2009) 1803-1810.

[13] A. Bohlin, E. Nordström, H. Carlsson, X.-S. Bai, P.-E. Bengtsson, Pure rotational CARS measurements of temperature and relative O₂-concentration in a low swirl turbulent premixed flame, *Proceedings of the Combustion Institute* 34 (2013) 3629-3636.

[14] A.C. Eckbreth, *Laser diagnostics for combustion temperature and species*, CRC Press 1996.

[15] S. Roy, J.R. Gord, A.K. Patnaik, Recent advances in coherent anti-Stokes Raman scattering spectroscopy: Fundamental developments and applications in reacting flows, *Progress in Energy and Combustion Science* 36 (2010) 280-306.

[16] E.H. Van Veen, D. Roekaerts, On the accuracy of temperature measurements in turbulent jet diffusion flames by coherent anti-Stokes-Raman spectroscopy, *Combustion Science and Technology* 175 (2003) 1893-1914.

[17] S.P. Kearney, K. Frederickson, T.W. Grasser, Dual-pump coherent anti-Stokes Raman scattering thermometry in a sooting turbulent pool fire, *Proceedings of the Combustion Institute* 32 (2009) 871-878.

[18] P. Magre, P. Moreau, G. Collin, R. Borghi, M. Péalat, Further studies by CARS of premixed turbulent combustion in a high velocity flow, *Combustion and flame* 71 (1988) 147-168.

[19] J.D. Garman, D. Dunn-Rankin, Spatial averaging effects in CARS thermometry of a nonpremixed flame, *Combustion and Flame* 115 (1998) 481-486.

[20] T. Seeger, M.C. Weikl, F. Beyrau, A. Leipertz, Identification of spatial averaging effects in vibrational CARS spectra, *Journal of Raman Spectroscopy* 37 (2006) 641-646.

- [21] J.Y. Zhu, D. Dunn-Rankin, CARS thermometry in high temperature gradients, *Applied Physics B* 56 (1993) 47-55.
- [22] I.G. Shepherd, F.M. Porter, D.A. Greenhalgh, Spatial resolution effects of CARS in turbulent premixed combustion thermometry, *Combustion and Flame* 82 (1990) 106-109.
- [23] T. Parameswaran, D.R. Snelling, Estimation of spatial averaging of temperatures from coherent anti-Stokes Raman spectroscopy, *Applied optics* 35 (1996) 5461-5464.
- [24] R.D. Hancock, F.R. Schauer, R.P. Lucht, R.L. Farrow, Dual-pump coherent anti-Stokes Raman scattering measurements of nitrogen and oxygen in a laminar jet diffusion flame, *Applied Optics* 36 (1997) 3217-3226.
- [25] K. Frederickson, S.P. Kearney, A. Luketa, J.C. Hewson, T.W. Grasser, Dual-pump CARS measurements of temperature and oxygen in a turbulent methanol-fueled pool fire, *Combustion Science and Technology* 182 (2010) 941-959.
- [26] G. Magnotti, A.D. Cutler, P.M. Danehy, Development of a dual-pump coherent anti-Stokes Raman spectroscopy system for measurements in supersonic combustion, *Applied optics* 52 (2013) 4779-4791.
- [27] M.P. Thariyan, V. Ananthanarayanan, A.H. Bhuiyan, S.V. Naik, J.P. Gore, R.P. Lucht, Dual-pump CARS temperature and major species concentration measurements in counter-flow methane flames using narrowband pump and broadband Stokes lasers, *Combustion and Flame* 157 (2010) 1390-1399.
- [28] S. Roy, T.R. Meyer, M.S. Brown, V.N. Velur, R.P. Lucht, J.R. Gord, Triple-pump coherent anti-Stokes Raman scattering (CARS): temperature and multiple-species concentration measurements in reacting flows, *Optics communications* 224 (2003) 131-137.
- [29] S. Roy, T.R. Meyer, R.P. Lucht, V.M. Belovich, E. Corporan, J.R. Gord, Temperature and CO₂ concentration measurements in the exhaust stream of a liquid-fueled combustor using dual-pump coherent anti-Stokes Raman scattering (CARS) spectroscopy, *Combustion and Flame* 138 (2004) 273-284.
- [30] R. Lucht, V. Velur, G. Fiechtner, C. Carter, J.K. Grinstead, J. Gord, P. Danehy, R. Farrow, Measurements of temperature and CO₂ concentration by dual-pump coherent anti-Stokes Raman scattering, 39th Aerospace Sciences Meeting and Exhibit, Reno, Nevada, 2001.
- [31] A.D. Cutler, E.C.A. Gallo, L.M.L. Cantu, WIDECARS spectra fitting in a premixed ethylene-air flame, *Journal of Raman Spectroscopy*, (2015).
- [32] S.A. Filatyev, M.P. Thariyan, R.P. Lucht, J.P. Gore, Simultaneous stereo particle image velocimetry and double-pulsed planar laser-induced fluorescence of turbulent premixed flames, *Combustion and flame* 150 (2007) 201-209.
- [33] A. Eckstein, P.P. Vlachos, Digital particle image velocimetry (DPIV) robust phase correlation, *Measurement Science and Technology* 20 (2009) 055401.
- [34] H. Pitsch, L.D. De Lageneste, Large-eddy simulation of premixed turbulent combustion using a level-set approach, *Proceedings of the combustion institute* 29 (2002) 2001-2008.
- [35] O. Colin, F. Ducros, D. Veynante, T. Poinso, A thickened flame model for large eddy simulations of turbulent premixed combustion, *Physics of Fluids* (1994-present) 12 (2000) 1843-1863.
- [36] S.H. Won, B. Windom, B. Jiang, Y. Ju, The role of low temperature fuel chemistry on turbulent flame propagation, *Combustion and Flame* 161 (2014) 475-483.
- [37] A. Lipatnikov, *Fundamentals of premixed turbulent combustion*, CRC Press 2012.
- [38] N.L. Carr, R. Kobayashi, D.B. Burrows, Viscosity of hydrocarbon gases under pressure, *Journal of Petroleum Technology* 6 (1954) 47-55.

- [39] A. Satija, R.P. Lucht, Development of a combined pure rotational and vibrational coherent anti-Stokes Raman scattering system, *Optics letters* 38 (2013) 1340-1342.
- [40] M. Pealat, M. Lefebvre, J.P. Taran, P.L. Kelley, Sensitivity of quantitative vibrational coherent anti-Stokes Raman spectroscopy to saturation and Stark shifts, *Physical Review A* 38 (1988) 1948.
- [41] R.E. Palmer, The CARSFT computer code calculating coherent anti-Stokes Raman spectra: User and programmer information, Sandia National Labs., Livermore, CA (USA), 1989.
- [42] A.D. Cutler, G. Magnotti, CARS spectral fitting with multiple resonant species using sparse libraries, *Journal of Raman Spectroscopy* 42 (2011) 1949-1957.
- [43] A. Satija, S. Yuan, S.V. Naik, R.P. Lucht, Vibrational CARS thermometry and one-dimensional numerical simulations in $\text{CH}_4/\text{H}_2/\text{air}$ partially-premixed flames, *International Journal of Hydrogen Energy* 40 (2015) 6959-6969.
- [44] J.J. Charonko, P.P. Vlachos, Estimation of uncertainty bounds for individual particle image velocimetry measurements from cross-correlation peak ratio, *Measurement Science and Technology* 24 (2013) 065301.
- [45] A.N. Lipatnikov, J. Chomiak, Turbulent flame speed and thickness: phenomenology, evaluation, and application in multi-dimensional simulations, *Progress in Energy and Combustion Science* 28 (2002) 1-74.
- [46] A. Lipatnikov, Premixed turbulent flame as a developing front with a self-similar structure, in: S.Z. Jiang (Ed.), *Focus on Combustion Research*, Nova Science, New York, 2006, pp. 89–141.
- [47] I.G. Shepherd, Flame surface density and burning rate in premixed turbulent flames, *Symposium (International) on Combustion* 26 (1996) 373-379.
- [48] J.F. Driscoll, Turbulent premixed combustion: Flamelet structure and its effect on turbulent burning velocities, *Progress in Energy and Combustion Science* 34 (2008) 91-134.
- [49] H. Kobayashi, K. Seyama, H. Hagiwara, Y. Ogami, Burning velocity correlation of methane/air turbulent premixed flames at high pressure and high temperature, *Proceedings of the combustion institute* 30 (2005) 827-834.
- [50] S.S. Shy, Y.C. Chen, C.H. Yang, C.C. Liu, C.M. Huang, Effects of H_2 or CO_2 addition, equivalence ratio, and turbulent straining on turbulent burning velocities for lean premixed methane combustion, *Combustion and Flame* 153 (2008) 510-524.
- [51] J.F. Driscoll, A. Gulati, Measurement of various terms in the turbulent kinetic energy balance within a flame and comparison with theory, *Combustion and Flame* 72 (1988) 131-152.
- [52] Y.C. Chen, R.W. Bilger, Experimental investigation of three-dimensional flame-front structure in premixed turbulent combustion: II. Lean hydrogen/air Bunsen flames, *Combustion and Flame* 138 (2004) 155-174.
- [53] K.N.C. Bray, P.A. Libby, J.B. Moss, Unified modeling approach for premixed turbulent combustion—Part I: General formulation, *Combustion and flame* 61 (1985) 87-102.
- [54] S. Roy, T.R. Meyer, R.P. Lucht, M. Afzelius, P.-E. Bengtsson, J.R. Gord, Dual-pump dual-broadband coherent anti-Stokes Raman scattering in reacting flows, *Optics letters* 29 (2004) 1843-1845.

Table 1 Operating conditions for the CARS measurements

Flame #	1	2	3
Reynolds number		10000 ± 50	
Adiabatic Temperature (K)		2030 ± 50	
Equivalence ratio	0.80 ± 0.02	0.84 ± 0.02	0.89 ± 0.02
CO ₂ % by total mass	0.0	5.0 ± 0.1	10.0 ± 0.1
CH ₄ mass flow rate (mg/s)	111 ± 2	110 ± 2	109 ± 2
Air mass flow rate (mg/s)	2440 ± 20	2300 ± 20	2150 ± 20
CO ₂ mass flow rate (mg/s)	0.00	124 ± 2	246 ± 4
Pilot H ₂ mass flow rate (mg/s)		2.7 ± 0.03	
Pilot H ₂ heat release percent of total (%)		6	
Lewis number	0.99	0.98	0.97
Laminar flame speed (cm/s)	33.7	29.7	24.9
Laminar flame thermal thickness (μm)	70	80	90

RMS turbulence fluctuation (m/s)	1.7
Integral length scale (mm)	1

Table 2 Parameters for dimensionless distance calculation

Mean flame brush location	x/D or r/D at $C_{mean} = 0.5$	Mean flame brush thickness, δ_t/D
0% CO ₂ r/D=0	2.9	1.4
5% CO ₂ r/D=0	3.4	2.2
10% CO ₂ r/D=0	3.8	2.5
0% CO ₂ x/D=1.94 (r<0)	-0.32	0.56
0% CO ₂ x/D=1.94 (r>0)	0.45	0.46
10% CO ₂ x/D=1.94 (r<0)	-0.42	0.39
10% CO ₂ x/D=1.94 (r>0)	0.39	0.39

Fig. 1. Cross-sectional schematic diagram of the PARAT burner with dimensioned zoom-in views of important aspects.

Fig. 2. Schematic diagrams of the turbulent generators applied in the PARAT burner. The plates are 3.175 mm thick. All units are expressed in mm. (a) lower, (b) upper.

Fig. 3. Dual-pump vibrational CARS single shot spectra in flame 1 recorded at $r/D=0.0$ and $x/D=5.17$ above the burner exit.

Fig. 4. (a) Mean velocity (solid symbols), RMS velocity fluctuation values of axial velocity (empty), and total turbulence fluctuation values (cross symbols) and (b) turbulence intensity (solid symbols) and integral length scales (empty symbols) at $x/D=0.2$ for flames with 0%, 5%, and 10% CO₂ addition. Mean and RMS fluctuation values are computed from 1000 image pairs.

Fig. 5. Mean temperature (solid symbols) and RMS temperature fluctuation (empty symbols) values along the burner centerline for turbulent premixed flames with CO_2 addition of 0%, 5%, and 10%. (Repeated experimental results are shown in green symbols). Mean and RMS fluctuation values are computed from 1000 single shots.

Fig. 6. Mean temperature (solid symbols) and RMS temperature fluctuation (empty symbols) values along the radial direction for turbulent premixed flames with CO_2 addition of 0% and 10%. Mean and RMS fluctuation values are computed from 1000 single shots. Dashed lines indicate the burner centerline.

Fig. 7. Mean progress variable variance along the flame centerline and along a radius at $x/D=1.94$ for flames with different levels of CO₂ addition.

Fig. 8. Combustion intensity for flames with 0%, 5%, and 10% CO₂ addition.

Fig. 9. Comparison of RMS temperature fluctuation values as a function of mean temperature from a purely wrinkled flame theory (line) with CARS temperature measurements (symbols) in the flames in this study. (R) in parentheses for two of the flames indicate data from repeated experiments.

Flame 1 (0% CO₂)

Flame 2 (5% CO₂)

Flame 3 (10% CO₂)

Fig. 10. Temperature PDFs along the burner centerline of flame 1 (left), flame 2 (middle), and flame 3 (right).

Fig. 11. Temperature PDFs along the radial direction in flame 1 (left) and flame 3 (right).

Fig. 12. Correlation plots for single-shot measurements of temperature and CO₂ mole fraction for flame 1.

Fig. 13. Correlation plots for single-shot measurements of temperature and O_2 mole fraction for flame 1.