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Dual-Pump CARS Temperature and Species Concentration Measurements in a Supersonic Combustor

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ABSTRACT

The dual-pump coherent anti-Stokes Raman scattering (CARS) method was used to measure temperature and the mole fractions of N₂ and O₂ in a supersonic combustor. Experiments were conducted in NASA Langley Research Center’s Direct Connect Supersonic Combustion Test Facility. In this facility, H₂ and oxygen-enriched air burn to increase the enthalpy of the simulated air test gas. This gas is expanded through a Mach 2 nozzle and into a combustor model consisting of a short constant-area section followed by a small rearward-facing step and another constant-area section. At the end of this straight section, H₂ fuel is injected at Mach 2 and at a 30° angle with respect to the freestream. One wall of the duct then expands at a 3° angle for over 1 meter. The ensuing combustion is probed optically through ports in the side of the combustor. Dual-pump CARS measurements were performed at the facility nozzle exit and at four planes downstream of fuel injection. Maps are presented of the mean temperature, as well as N₂ and O₂ mean mole fraction fields. Correlations between fluctuations of the different measured parameters are also presented.

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INTRODUCTION

Supersonic combustion involves a complex interaction between turbulent mixing and chemical reaction. Whether efficient supersonic combustion occurs depends strongly upon the rate and extent to which chemical reactions occur compared to the residence time of the fluid in the combustor. In addition, no current ground testing facility is capable of perfectly reproducing the conditions of a real combustor in flight, due to the excess water (and often also carbon dioxide) in combustion-vitiated facilities, and the vibrational nonequilibrium, excess nitric oxide and atomic species, and short test times of pulsed facilities. If all these effects are to be accounted for, it is important to have available non-intrusive and spatially resolved methods for determining the location and extent of chemical reaction within an engine.

Modeling fuel-air mixing and combustion using computational fluid dynamics (CFD) requires quantitative data from within the flow to evaluate the performance of turbulent mixing models and to develop new models. Although measurements of surface properties such as pressure and heat transfer are commonplace, quantitative or semi-quantitative measurements of flowfield properties are difficult to obtain, and hence there are far fewer examples in the literature. Non-intrusive measurements have been performed in supersonic mixing and combustion experiments using coherent anti-Stokes Raman scattering (CARS), laser-induced fluorescence (LIF), and other techniques. While CARS can obtain point-wise multi-parameter measurement of flow properties for major species such as N$_2$ and O$_2$, LIF provides spatially-resolved flowfield visualization and measurements of temperature and mole fraction for minority species. Information about OH, CH and other radical species provided by LIF is particularly useful in understanding combustion interactions. LIF is also easily extended to planar measurements, allowing instantaneous and high-spatial-resolution data to be acquired. However, LIF has not been used to produce quantitative measurements of mole fraction for majority species such as H$_2$, N$_2$ and O$_2$ in combusting flow. Thus, CARS and LIF are complementary measurement techniques.

Some forms of the CARS technique are particularly valuable in performing quantitative studies of supersonic combusting flows because they have the potential to measure temperature and the concentrations of multiple important combustion species simultaneously. Other techniques such as Raman scattering can measure the same quantities, but CARS has the advantage of producing a coherent signal beam. This increases the signal-to-noise ratio of measurements and permits spatial filtering to reduce the degradation of measurement quality due to
interference from flow luminosity. The coherent nature of the CARS signal also means that good signal-to-noise ratio measurements can be made where optical access to the flow is limited, which is the case for scramjet combustors. When flow properties are measured simultaneously, correlations between the different species mole fractions and between the temperature and species mole fractions can be measured. These correlations can then be used to evaluate parameters in proposed turbulence models, or to determine the effectiveness of large eddy simulation turbulence models.

There are several varieties of the CARS technique, and the choice of a particular method has a significant effect upon the difficulty and results of a given supersonic combustion experiment. Conventional broadband N₂ CARS is ubiquitous and has proved very useful for providing temperature measurements in a number of supersonic combustion studies,¹,²,⁶,⁷ but it is more difficult to make mole fraction measurements.¹² Because mole fractions of the other species are not generally known, their contribution to nonresonant susceptibility will also not be known, and this quantity is required for an absolute measurement of mole fraction.

In order to obtain more information about composition of the combusting flow, various multi-species techniques have been developed. The three most used methods are dual-Stokes CARS, pure-rotational CARS and dual-pump CARS.¹² Dual-Stokes CARS, in which two broadband beams interact with two pump beams to produce two spatially and spectrally separated CARS signals simultaneously, has been used for multi-species measurements.⁵,¹³,¹⁴ This system requires accurate measurement of all three laser intensities and calibration of multiple detectors to make quantitative measurements of mole fractions. Pure rotational CARS¹⁵,¹⁶ is another variation that measures multiple species. The pure rotational CARS spectra for all species occur in the same spectral region, which is very useful for multi-species measurements, although for the same reasons pure rotational spectra can become very complex, making it difficult to separate spectral contributions from different species. In general, rotational CARS is more sensitive at low temperatures, but vibrational CARS spectra are more sensitive to temperature variations at flame temperatures.¹⁷

In this paper, we use the dual-pump CARS method developed by Lucht and co-workers¹⁸,¹⁹ and extended by us, to simultaneously measure temperature and the absolute mole fractions of N₂ and O₂ in a supersonic combustion experiment. This experiment followed on from an initial experiment at the same nominal conditions in which conventional broadband N₂ CARS was used to map the temperature field in a supersonic combustor.¹ Using dual-
pump CARS in the current experiment allowed the temperature field and species mole fraction fields to be simultaneously measured. Like pure-rotational CARS, dual-pump CARS allows the CARS signals from multiple species to appear in the same spectral region so that they can be captured on a single detector. Since the same three beams are used in the excitation process for all species, relative beam energies need not be measured, nor do multiple detectors need to be calibrated. Unlike pure-rotational CARS, dual-pump N₂-O₂ CARS provides some flexibility in the spectral location of the O₂ signal relative to the N₂ signal, minimizing overlaps between the spectra. Furthermore, the relative intensity of the O₂ and N₂ CARS signals is unaffected by beam steering, an important consideration in supersonic combustion flows. This supersonic combustion experiment is complemented by an analogous non-reacting case in which the same dual-pump CARS system has been used to study fuel/air mixing at the same nominal freestream conditions.²⁰

Conventional broadband N₂ CARS uses two spectrally narrow green beams as pump beams and one spectrally broad red beam as the Stokes beam. The frequency difference between the green and red beams typically corresponds to the vibrational Raman shift of N₂. The CARS signal is then a faint, spectrally broad blue beam that contains the N₂ spectrum. This spectrum can be detected and then fitted with a theoretical model on a computer to determine the temperature. The dual-pump CARS technique used here differs from the conventional broadband N₂ CARS technique because it uses two different colors for the pump beams: one green and the other yellow. The same broadband red beam is used as in conventional broadband N₂ CARS. The frequency of the yellow pump beam is chosen so that the frequency difference between the yellow and red beams excites the vibrational Raman transitions of O₂, with the green and red beams exciting the vibrational Raman transitions of N₂. The resulting blue CARS spectrum contains both N₂ and O₂ spectra. The relative intensities of these two spectra provide a measure of the relative mole fractions of N₂ and O₂.

Coincidentally, several pure-rotational Raman transitions of H₂ are present in these spectral regions as well, as described in Refs. 20 and 21. These H₂ transitions were measured in the present experiment, in principle allowing the relative mole fractions of N₂, O₂ and H₂ to be quantified simultaneously. However, comparisons between calculations and measurements of H₂ mole fraction in an atmospheric pressure flame showed that the measured H₂ mole fraction was too low by 10–15% compared to the predictions from an adiabatic flame calculation, perhaps owing to errors in our spectral lineshape modeling. Thus, the H₂ measurements have been excluded from the current
paper. Recently, others have also simultaneously measured N₂, O₂ and H₂ in a flame using triple-pump CARS, but the system described here has the advantage of requiring one less laser and therefore being simpler to implement. In this paper we also demonstrate that applying N₂-O₂-H₂ dual-pump CARS to H₂-air combustion has an important advantage: simultaneous measurement of CARS signal from three out of the four major species provides sufficient information about the gas composition, and hence the contributions of the major species to the nonresonant susceptibility, to allow absolute N₂ and O₂ mole fractions to be determined. During previous use, dual-pump CARS has only allowed determination of ratios of species mole fractions, such as CO₂/N₂ ratio.

COMBUSTOR MODEL AND FLOW FACILITY

The experimental facility, model, and operating conditions are nominally identical to those tests described in Ref. 1, although the present tests were conducted some two years after. Many of the method descriptions herein are similar to those of Ref. 1, but have been repeated herein for convenience of the reader.

The experiment was conducted in NASA Langley’s Direct-Connect Supersonic Combustion Test Facility (DCSCTF). The facility is designed to test the combustor of a scramjet engine by directly connecting the facility nozzle exit to the entrance of the combustor. The stagnation enthalpy of the gas is increased by burning H₂ in air and replenishing the O₂ lost in the combustion. The nozzle gas flow rates are selected so that the mass fraction of O₂ is the same as that of standard air. This high-pressure, vitiated gas is accelerated through a water-cooled convergent-divergent Mach 2 nozzle, before entering the test model.

The nominal test conditions are nearly identical to those presented in a previous single-pump CARS study, and are representative of Mach 7 flight. Gas flow rates to the heater are: 0.916 ±0.008 kg/s air, 0.0284 ±0.0006 kg/s H₂, and 0.302 ±0.005 kg/s O₂. The heater stagnation pressure is 0.765±0.008 MPa. All uncertainties presented in this paper are on the 95% confidence interval half widths (CIHW), equivalent to 1.96 times the standard deviation divided by the square-root of the number of samples. The above uncertainties are due to the random run-to-run variations and do not include ±3% uncertainty in the mass flow rate measurements.

Heater and nozzle exit conditions were estimated in Ref. 1 from the flow rates, heater pressure, and nozzle minimum and exit areas using one-dimensional (1D) analysis detailed in Ref. 24 and assuming equilibrium chemistry. The nominal calculated conditions, and uncertainties due to mass flow rate measurement error and run-
to-run variations in heater conditions are: heater stagnation temperature $1818\pm75$ K, exit temperature $1181\pm60$ K, mole fraction $O_2$ 0.186, mole fraction $N_2$ 0.512, exit pressure $100\pm1.5$ kPa, exit Mach number $1.989\pm0.005$. Errors arising from the assumption of 1D flow (the effects of non-uniform composition, boundary layers, etc.) are assumed to be small. A study of the flow quality at the exit of the facility nozzle was previously conducted and is described in Ref. 25.

![Figure 1](image)

**Figure 1.** (a) Schematic of scramjet combustor model. (b) Close-up of injection region. (From Ref. 1.)

The test model, known as the Scholar model, is shown in Fig. 1; flow direction is from left to right. The model consists of two main duct sections: a copper upstream section and a carbon steel downstream section. Stainless steel flanges and carbon gaskets separate the sections from each other and from the nozzle. Proceeding from left to right, there is a Mach 2 nozzle, a constant area segment, a small outward step at the top wall, a second short constant-area segment and a segment with constant $3^\circ$ divergence of the top wall. The span is constant at 87.88 mm. The fuel injector is located immediately downstream of the start of the $3^\circ$ divergence. The injection angle is $30^\circ$ to the opposite wall. The injector nozzle is designed by the method of characteristics to produce Mach 2.5, one-dimensional flow at the injector exit. Hydrogen is injected at a stagnation pressure of $3.44\pm0.065$ MPa** and a

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** In Ref. 1, the stagnation pressure of primary injected $H_2$ was incorrectly reported as 2.12 MPa.
stagnation temperature of 302±4 K. This fuel flow rate corresponds to an overall equivalence ratio of 0.99±0.04 for the gas in the duct.

The duct is not actively cooled; however, the wall thickness of the copper duct is greater than 32 mm and the carbon steel duct is 19 mm thick. Thus, given the good thermal conductivity of these materials, it is possible to operate the facility with the model fueled for run times in excess of 20 seconds without reaching excessive temperatures. With atmospheric-temperature air flowing in the model between runs, the tests can be repeated every 10 to 15 minutes. A total of 15 to 25 runs could be obtained per day.

The model is equipped with 7 pairs of slots, to allow the CARS beams to enter and exit the duct. The first slot is at the nozzle exit and the other 6 slots are downstream of the fuel injection, as shown in Fig. 1. Slots 1, 3, 5, 6, and 7 were used in this study. The slots are used in pairs, one on each side of the duct. They are 4.8 mm wide and extend the full height of the duct. When not in use, the slots are plugged flush to the duct wall. Windows covering the slots are mounted at the end of short rectangular tubes at the Brewster angle, to minimize reflections. The tubes connecting the windows and combustor are ventilated with a constant flow of electrically heated (~400 K) dry air to prevent condensation of water vapor from the facility on the windows. The CARS interaction region (the measurement volume) can be translated the full span and height of the duct without damaging the windows. The supersonic combustor model is also instrumented with pressure taps and wall temperature probes, and measurements using these probes have been presented previously for the same configuration and flow conditions as the present experiment.

**CARS EXPERIMENTAL SYSTEM**

A complete description of the dual-pump CARS system is given in Ref. 26. It used an unseeded frequency-doubled Nd:YAG laser (Spectra Physics DCR-4), producing about 550 mJ per pulse at 532 nm with ~1 cm⁻¹ line width. The pulse duration was 10 ns and the repetition rate was 10 Hz. The output of the YAG laser was split three ways. Approximately 80 mJ was used as the green pump beam for CARS. A purpose-built, axially pumped broadband dye laser operating at 607 nm was pumped using 250 mJ of green light from the doubled Nd:YAG. The dye solution consisted of rhodamine 640 laser dye in methanol, and the dye laser energy was 20–25 mJ. The line shape of the broadband laser was well represented by the sum of two Gaussian functions. The “double” Gaussian
was necessary to ensure proper modeling of the wings of the spectrum, important since the CARS spectrum can include strong H\textsubscript{2} lines near the edge of the profile.

Another 200 mJ of 532-nm light was used to pump a yellow (554-nm) narrowband dye laser (Lambda Physik, FL3002, which was modified according to the manufacturer's instructions from excimer to YAG pumping), producing 25 mJ of yellow light. The green and red beams were combined using a dichroic mirror. These two beams and the yellow beam were then passed through a spherical lens having a focal length of 410 mm. The planar BoxCARS phase-matching geometry was used, with the green and red beams overlapping. The probing volume formed at the intersection of the three beams had a minimum diameter of 130 µm, full width at half maximum (FWHM), measured by traversing a knife-edge across the foci of all three beams and measuring unblocked light. The probe volume was 1.8 mm long (FWHM), measured by traversing the probe volume across a thin planar jet of N\textsubscript{2} gas, surrounded by a 30-mm-diameter jet of argon, and measuring the N\textsubscript{2} CARS signal as the probing volume was traversed across the N\textsubscript{2} sheet.

A second 410-mm focal-length lens collected and collimated the three input beams and the 491-nm CARS signal beam. Two dichroic mirrors, efficiently reflecting blue light while transmitting yellow and green light, were arranged in a multi-reflection configuration to isolate the blue signal beam. This blue beam was then directed into a one-meter focal-length spectrometer (McPherson) with a 1200-groove/mm grating. Two cylindrical lenses provided a sharp horizontal focus at the spectrometer entrance that was imaged, spectrally dispersed, at the detector. The detector was an 1100×330-pixel, non-intensified, back-illuminated CCD camera (Pixel Vision, SV11CBJ). A LabVIEW interface downloaded the spectra to a PC for subsequent analysis and could display spectra in real time for signal optimization. The 330-pixel region was binned into three separate rows of 110 pixels before being read out, to allow more rapid acquisition of data. This resulted in three 1100-pixel spectra per laser pulse, with a spectral resolution of 0.74 cm\(^{-1}\) per pixel. The CARS signal was imaged onto the first two of these regions, with the contributions of dark noise and laser scatter acquired on the third bin region. Distributing the signal over two bin regions increased the dynamic range of the measurements by up to a factor of two. The binned background was subtracted from each of the signal bin regions before the signal regions were summed to form the total spectrum.

The spectral non-uniformity of the broadband Stokes laser was accounted for by normalizing the CARS spectrum to the “non-resonant” CARS spectrum obtained immediately after completion of the day's runs. The non-
resonant CARS spectrum, which varies with the profile of the broadband laser, was measured by obtaining a CARS spectrum in a flow of argon (which has no resonances in this range of wavelength), averaged over 200 laser pulses. A sum of two Gaussian functions was fitted to the non-resonant CARS spectrum, as mentioned above, as this fits the wings of the non-resonant CARS spectrum much better than the single Gaussian function used in Ref. 1. The full-width at half-maximum of the non-resonant spectrum was approximately 130 cm\(^{-1}\) in the spectral region of the CARS signal imaged by the camera. This was sufficiently broad to contain spectral features of all three species of interest. The fitted function was used for the normalization.

**CARS SPECTRAL DATA ANALYSIS**

The analysis of the CARS spectra differs in several important ways from the analysis reported in our previous paper,\(^1\) due to both the wider spectral region being fitted and to the larger number of fit parameters. The most important changes will now be discussed.

The broadband Stokes laser spectrum varies during a day’s facility runs by as much as 30 cm\(^{-1}\), due to alignment and temperature fluctuations in the optical system. Because important spectral features occur in the wings of the non-resonant spectrum where normalization causes signal levels to be particularly sensitive to changes in the spectral location of the Stokes beam, the analysis must account for these changes. This was done by allowing the non-resonant CARS spectrum to shift in wavelength, while maintaining spectral width and shape, as part of the fitting process. For each tunnel run, the middle one-third of acquisitions was fitted for temperature and the three mole fractions using the non-resonant spectrum acquired in argon gas before the day’s facility runs. The same spectra were then fitted twice more using the same non-resonant spectrum shifted by 7.4 cm\(^{-1}\) above and below the nominal spectral location. A systematic spectral offset in the center of the Stokes spectrum manifests as a gradient in the non-resonant CARS signal. The most correct of the Stokes spectra produces the smallest residual in the fitted spectra. If the middle of the three Stokes spectra used for normalizing the tunnel CARS spectra did not have the minimum residual, the center of the Stokes beam spectrum would be shifted another 7.4 cm\(^{-1}\) in the direction of decreasing residual until a minimum was found. A parabola was fitted to the residual at these three lowest-residual points and the minimum of that parabola was used as the location of the center of the Stokes beam spectrum for all the fits in that facility run. This procedure was repeated for all the facility runs on each day, and determined the shift in the Stokes laser spectrum to within 3.5 cm\(^{-1}\), or 2.7% of the Stokes laser's FWHM.
Individual measured CARS transitions are spectrally broader than theoretical transitions, mainly due to the low-resolution dispersion of the spectrometer. The shape and width of this broadening, known as the “instrument function”, was determined by fitting room-temperature air spectra on each day tunnel data was acquired. The width and amplitude of the instrument function were varied using a nonlinear least-squares algorithm to produce the best fit for the known room-temperature air parameters. This fitted instrument function was used to generate the library of theoretical spectra for that day’s runs. A version of the CARSFIT code, originally developed at Sandia National Laboratories, Livermore, CA, and modified for the dual-pump CARS technique by Hancock and coworkers, was used to compute the theoretical CARS spectra.

In our previous paper, temperature and N₂ mole fraction were fitted for each measured CARS spectrum by computing spectra at a large number of discrete temperature and mole fraction values. Once this database of spectra had been calculated, the least-squares best fit was determined by comparison with the measured spectrum. The measurements in this paper could not be fit with the same resolution, because there were two additional fit parameters, the mole fractions of O₂ and H₂, and because the fitting of the very narrow H₂ transitions required higher spectral resolution in the library. In order to quickly fit the many thousands of CARS spectra obtained in the experiment, CARSFIT was used to generate a coarse 4-dimensional (temperature, N₂, O₂, and H₂ mole fractions) library of spectra, using the Voigt profile to model the individual Raman transition strengths and assuming that the only other gas present was H₂O. No attempt was made to account for the concentrations of minor species such as OH, NO or Ar. We modified the CARSFIT code to include convolutions with a second pump beam, as the previous version of the code assumed one of the pump beams to be monochromatic. The Nd:YAG laser used in these experiments was not injection-seeded, so the laser line width is large compared to the line width of the individual Raman transitions. We also removed from the code an error in the adaptive grid calculation, allowing the faster adaptive grid calculation to be used for H₂ spectrum calculations.

The measured N₂, O₂ and H₂ mole fractions were used to estimate the nonresonant susceptibility. This was calculated for each measurement using the fitted gas composition and assuming that the only other major species present was water vapor. Having a reasonable estimate of the contributions of the major species to the nonresonant susceptibility allows quantitative absolute measurements of N₂ and O₂ mole fractions.
The library of theoretical CARS spectra contains temperature intervals of 200 K between 200 K and 3000 K, and between 10 and 12 mole fraction values, depending upon the species. Oxygen mole fractions are included between 0 and 0.4, while the N₂ and H₂ mole fractions vary between 0 and 1. Mole fraction values are more closely spaced near zero, to achieve better resolution at low molar concentrations. Even this coarse grid of computed spectra required 60 hours to generate on a 2.4 GHz DEC alpha, with a different database required for each day’s runs due to variations in the instrument function from one day to the next. An initial guess of temperature and composition is obtained by comparing each measured spectrum to every theoretical spectrum in this coarse library to find a best fit. A more precise estimate of temperature and composition is then obtained using “theoretical” spectra linearly interpolated between the library entries: the absolute deviation between these theoretical spectra and the measured spectrum was minimized using a nonlinear least-squares method. Errors caused by this interpolation scheme were far smaller than other random errors in the experimental data: a set of thirteen test spectra generated by CARSFIT over the expected temperature and mole fraction ranges for this experiment were fitted with average temperature errors of 2.1 K and mole fraction errors of less than 0.003. The largest systematic errors in fitted temperature and mole fraction for this test data were 20 K and 0.008 respectively. Both these values are significantly smaller than the uncertainties in the measurements for the laminar flat flame experiments described subsequently.

**MDOE AND RESPONSE SURFACE METHODS**

The experimental data were obtained using a formal experiment design method, known as modern design of experiments (MDOE). This method, first applied in the analysis of planes of CARS data in Ref. 1, with more details in References 28 and 29, is briefly summarized here. The CARS probe volume was scanned through the flowfield along horizontal and vertical lines – one scanned line per tunnel run. The order and direction of these scans was randomized to defend against systematic errors caused by factors like heating of the duct during a facility run. Repeat measurements were obtained to improve measurement uncertainty and to provide a qualitative determination of the uncertainty and repeatability of the measurement system and flow facility.

After the CARS spectra were processed to determine the temperature and the mole fractions at various locations in the duct, response surfaces were fitted to these data to provide quantitative maps of the mean flowfield parameters. For each plane, a response surface was fitted to the temperature and mole fraction data to determine
surfaces of the mean (time-averaged) value of these properties. Using these fitted mean surfaces then allows us to compute statistics for the fluctuations in the measured parameters.\textsuperscript{1,28,29} The difference between each measured sample and the mean, denoted by a primed quantity, such as $T'$ for temperature fluctuations and $\chi_i'$ for mole-fraction fluctuations of species $i$, and the chosen fluctuation product, such as $T'T'$, is calculated. The fluctuation product data are then fit to response surfaces, as described above, to determine surfaces of mean values of the product, such as $<T'T'>$. In this way, for example, the temperature variance, $<T'T'>$, and the root-mean-square (RMS) temperature fluctuation, $(<T'T'>)^{1/2}$, can be computed. Similarly, correlations between various fluctuating properties in the flow, such as $<T'\chi_{i2}>$ are found.

In the initial report of this experiment in Reference 21 and also in the analysis of a similar data set in Reference 20, response surface fits using the cosine series bivariate order 6 function\textsuperscript{30} were used but were too smooth to represent the sharp gradients in temperature and species concentration present in several data maps, particularly in the shear layer between the fuel jet plume and the air. To correct for this limitation, a “saturated Gaussian” function was added to the cosine series bivariate order 6 function in order to better represent these steep gradients caused by injection of the fuel plume into the combustor. This function, inspired by a saturated absorption spectrum, is given by:

$$G_{sat}(x, y) = a + b \left(1 - e^{-c\left(\frac{x}{d} - e\frac{y}{f}\right)^2}\right)$$

where $a$, $b$, $c$, $d$, $f$, $g$, and $h$ are free parameters and $e$ is the base of natural logarithms. For values of $c$ near unity, this function resembles a positive Gaussian function. As $c$ increases from unity, the edges of the Gaussian become steep and the top of the Gaussian becomes flat. The $b$ parameter scales the height of the Gaussian and, when negative, represents a decrease in the measured quantity. This functional shape duplicates that observed in the experimental temperature and species mole fraction maps close to the fuel plume with better fidelity than the cosine series bivariate order 6 function alone.

The terms with coefficients that are statistically insignificant are removed from the fit. The significance is determined by whether the statistical P-value for the term is greater than 5 percent and is therefore considered by statistical fitting standards to be found by chance.\textsuperscript{31} This removes terms that did not improve the quality of the fit,
thereby improving the confidence in the remaining parameters. Consequently, the confidence interval half width uncertainty, which depends on the number of parameters in the fit equation, is also reduced. Removing terms removes uncertainty added by terms that did not add significantly to the fidelity of the fit. These fits, by virtue of their limited number of fit coefficients (much less than the number of data points), smooth (or average) the data.

**CARS RESULTS**

_Furnace and Flame Calibrations_

(a)

![Temperature vs. Equivalence Ratio](image1)

(b)

![Mole Fraction vs. Equivalence Ratio](image2)

Figure 2. Hencken burner CARS calibration measurements of (a) temperature and (b) mole fractions of O₂, N₂ and H₂.
The CARS system was characterized by making measurements in an atmospheric-pressure furnace and a Hencken flat-flame burner. Mean values of temperature and mole fraction were obtained. Mean temperature measurements in a furnace containing air in approximately 250-K increments between 300 and 1800 K agreed with thermocouple measurements within 26 K on average, while the mean measured mole fractions of N\textsubscript{2} and O\textsubscript{2} agreed to within 1.6% of their nominal values for atmospheric air. The temperature measurement standard deviation averaged 64 K while the standard deviation of the species mole fractions averaged 7.8% of the measured value for O\textsubscript{2} and 3.8% of the measured value for N\textsubscript{2}, based on 200 single-shot measurements at each of the seven measured temperatures.

The measurements in a Hencken burner, which produced an atmospheric-pressure, adiabatic, H\textsubscript{2}-air laminar, flat flame at fuel-lean and fuel-rich conditions, are summarized in Fig. 2. Results are compared to an equilibrium, adiabatic flame calculation, and the bars on the plots indicate the standard deviation of 200 measurements. Temperatures in the flame were measured with a standard deviation of ~70 K over the entire temperature range. The 95% confidence interval half width (CIHW) uncertainty was ~10 K. The average absolute value of the difference between measured and calculated mean temperatures was 29 K, indicating systematic error sources. Mole fractions of N\textsubscript{2} and O\textsubscript{2} were measured with a standard deviation of 0.027 and 0.026 respectively, and the average absolute differences between the measurement and the calculation were 0.010 and 0.009. Errors in mole fractions were greater than expected based on random error. The standard deviation of the H\textsubscript{2} mole fraction measurements in the Hencken burner varied from 0.003 at the fuel-lean equivalence ratios to a maximum of 0.028 at the maximum equivalence ratio of 3.71.

Measured H\textsubscript{2} mole fractions in the flat flame were systematically low by 10-15% when compared to the computation. In Reference 21, the measured H\textsubscript{2} mole fraction also appeared to be low for the combustor data. Analysis of the combustor data, based upon the measured values of O\textsubscript{2} and N\textsubscript{2} mole fraction, indicates that the discrepancy is not constant, either with H\textsubscript{2} mole fraction or from one day’s runs to the next. These errors may be either a result of inadequate spectral modeling or a systematic error in the measurement of the H\textsubscript{2} signal. Due to the unresolved error in the H\textsubscript{2} mole fraction measurements, H\textsubscript{2} measurements in the combustor are not reported in this paper. Although the exact value of the fitted H\textsubscript{2} mole fraction is questionable, the combustor measurements produced qualitative variations in H\textsubscript{2} mole fraction that were consistent with expectations: the H\textsubscript{2} mole fractions
Figure 3. Sample CARS spectra from planes 5 and 6.
increased as the mixture became more fuel-rich. As mentioned previously, the measured H₂ mole fraction was used to calculate the mole fraction of H₂O, which determines the non-resonant susceptibility of the mixture for each measurement. The systematic error in H₂ mole fraction can be expected to have a secondary effect on the measured values of N₂ and O₂ mole fraction.

**Combustor Measurements**

![Temperature and mole fraction plots for a single facility run in plane 6.](image)

**Figure 4.** (a) Temperature and (b) mole fraction plots for a single facility run in plane 6.

Figure 3 shows several representative CARS spectra and fits measured in the combustor. These spectra show the quality of fits to the data for planes 5 and 6, at a variety of flow conditions characteristic of (a) the freestream
conditions, (b) hot flow containing H₂ and N₂, (c) cooler flow outside the duct in the window space and (d) cooler flow in the unburned fuel jet plume containing only small proportions of N₂ and O₂.

Figure 4 contains a set of measurements for a single tunnel run in which the CARS measurement volume was scanned horizontally across the duct at plane 6 at a fixed height. The co-ordinate system is chosen such that its origin is in the centre of the duct in the freestream plane, with the bottom wall of the duct at \( y = -19.8 \) mm. This plot was obtained at \( y = 13.7 \) mm, near the center of the fuel jet plume. The plot shows the scatter of the temperature and mole fraction measurements during the scan. Most of this scatter, particularly in the high-temperature portion of the plot, is due to turbulent variations in the conditions rather than measurement random error: the standard deviation of the CARS system is significantly smaller then the variations seen in the traverse. The cosine bivariate curve fit in Fig. 4 (a) does not fit the large temperature gradient perfectly near the duct wall, but adequately fits the mole fraction variations and the temperature in the middle of the duct. This poor fit near the duct wall is caused by the inability of this fitting function to capture steep gradients in the data set. Other functional forms were able to fit this steep data better near the wall, but this functional form was used because it is well behaved in regions of the flow where there is no data and for consistency with all the other data planes.

Figure 5. Cut-away view of data acquisition locations.
The measurement locations for each of the data points are shown in Fig. 5. Each plane of data produced 2000-3000 data points, depending upon the amount of time available for runs. Most measurement planes were obtained in a single day. Plane 7 required two days of tunnel runs to generate all the necessary data. The grids indicate horizontal and vertical traverses across the duct, while single points away from these grid lines indicate runs for which the measurement volume was not moved during the run. Several planes also contain repeat runs along a given line, performed to test the repeatability of the measurements for nominally identical conditions.

Figure 6 is a summary of the distributions of mean temperature and species concentration, measured by the CARS system and fitted with the response surfaces. At plane 1 the mean properties are essentially uniform; the temperature might be expected to change within the boundary layers, but these are not resolved by the measurements. Taking the average of all measurements across the duct between \( z = \pm 40 \text{ mm} \), the measured properties are \( T = 1250 \text{ K} \), \( \chi_{\text{O}_2} = 0.204 \) and \( \chi_{\text{N}_2} = 0.494 \) with standard deviations of 151 K, 0.024 and 0.042 respectively. As previously stated, the nozzle exit conditions were calculated to be \( T = 1181 \pm 60 \text{ K} \), \( \chi_{\text{O}_2} = 0.186 \) and \( \chi_{\text{N}_2} = 0.512 \). The measurements of temperature at plane 1 do overlap with the predicted value to within the CIHW of 25 K. The fact that the temperature standard deviation is more than twice the 70 K measured in the laminar flame indicates that the temporal variations in freestream temperature are large enough to be resolved by the CARS system and that there is substantial freestream turbulence. If we assume that the real temperature fluctuations are statistically independent of the random error in the temperature measurement, and hence add in quadrature, we estimate that the standard deviation of the plane 1 temperature (the RMS turbulent fluctuation level) is \( \approx 130 \text{ K} \).
discrepancies between the measurements of mean mole fraction and the calculations are similar in magnitude to the discrepancies between the measurements and calculations observed in the furnace and Hencken burner.

The temporal variations in the vitiated air flow away from the fuel jet was determined at plane 5 by placing the measurement volume at a fixed location (-42 mm, -6.8 mm) over 200 spectrum acquisitions. This location was away from both the fuel jet plume and the combustor wall. The mean flowfield properties over this run are $T = 944$ K, $\chi_{O_2} = 0.199$ and $\chi_{N_2} = 0.538$ with standard deviations of 114 K, 0.022 and 0.037 respectively. As expected, the freestream temperature in plane 5 is less than that of plane 1 because the freestream expands as the duct diverges downstream of injection. The standard deviations in temperature and mole fraction, expressed as a percentage of the mean value, are similar for both planes 1 and 5.

Another interesting outcome of the plane 1 measurements is the nonuniform distribution of both $O_2$ and, to a lesser extent on a percentage basis, $N_2$ mole fraction, with increasing concentrations of both going from left to right across the plane. This observation was consistent over two independent measurements in plane 1, and was also observed at different combustor entrance conditions in the experiments of Tedder et al. (2005)$^{12}$. This is not an artifact of the heating of the facility during each run, because care was taken to traverse the duct in both positive and negative $z$-directions to prevent such systematic errors. Similar asymmetries can also be seen near the edges of planes 5, 6 and 7, with the $O_2$ mole fraction always increasing from left to right. The systematic gradient in plane 1 $O_2$ concentration across the duct is approximately 0.03, greater than the average error in $O_2$ mole fraction for the flat flame measurements (which was 0.01). The plane 1 temperature map does not show an obvious gradient. These trends do not appear to be self-consistent (a composition variation would predict a temperature variation) nor consistent with the way the facility heater is constructed (axisymmetric); this discrepancy remains unresolved.

The temperature maps in Fig. 6 can be directly compared with the previous conventional broadband $N_2$ CARS measurements$^1$ obtained at the same nominal conditions. Upon comparing the temperature surface fits for the two campaigns, the temperatures averaged over the plane were found to differ by -97, -6, -95, 90 and -17 K in planes 1, 3, 5, 6 and 7 respectively. The greatest difference between the surface fits occurred in plane 1. The plane 1 average temperature in Ref. 1 is 1162 K, with CIHW of 30 K, while the average in plane 1 for the current series of experiments is 1250 K. More detailed comparisons show a systematic difference of 80–120 K throughout the central part of the combustor, with the dual-pump CARS producing the higher temperatures.
Initially it was thought that a possible reason for the discrepancy between the conventional and dual-pump CARS measurements in plane 1 was due to a leak in the nozzle cooling water system which developed during acquisition of the dual-pump CARS data, causing cooling water to enter the flow. Some preliminary dual-pump CARS measurements had been performed in plane 1 before the leak had been detected, allowing us to compare these two data sets to see if the average temperature had changed after the repair. The preliminary measurements were performed at the very beginning of the dual-pump CARS experimental campaign, when the combustor was configured exactly as it had been in the experiments of Ref. 1. Figure 7 compares two horizontal traverses at \( y = -1.8 \) mm: one obtained before the nozzle repair and another obtained afterwards and used for the plane 1 surface presented in Fig. 6. This plot plainly shows little difference between the measured temperatures for the two traverses. The plane-averaged temperatures agree to within 27 K, or 2.1\%, of one another. The agreement is similarly good for other traverses in the same region, and between the \( \text{N}_2 \) and \( \text{O}_2 \) mole fraction measurements in the two plane 1 dual-pump CARS experiments. One difference between the two measurements in Fig. 7 is that the 110-K standard deviation of the second series of measurements is noticeably smaller than the 140-K standard deviation in the first plane 1 measurement. This difference is due to a reduction in instrument random error derived from the ability to remotely align two of the pump beams between experiments (thereby increasing signal level). This capability was added in the period between the two measurements. Considering that the temperature measurements in Fig. 7 were obtained two months apart, with data for planes 3, 5, 6 and 7 acquired between the two measurements, the dual-pump CARS measurements and the facility combustor entrance flow are consistent.

\[\text{Figure 7. Data for facility runs obtained before and after nozzle repair.}\]
The reasons for the discrepancy between the dual-pump CARS temperature measurements and those of Ref. 1 for plane 1 are not known. The good agreement between the dual-pump measurements in the Hencken burner and adiabatic flame calculations, and a greater confidence in the spectral fitting process due to the presence of O$_2$ spectra as well as N$_2$ in our measurements, gives us more confidence in the plane 1 dual-pump CARS measurements than for the single-pump CARS measurements.

Returning to Fig. 6, the first strong indications of combustion occur at the top of the duct in plane 5, where the temperature increases to a peak value of approximately 1600-1700 K, compared to 800-900 K for the same location in the mixing-only experiment. This also corresponds to a region of reduction of the O$_2$ to N$_2$ ratio above the center of the fuel plume, indicating consumption of O$_2$ by combustion. The fact that combustion is already occurring in plane 5 is of interest, because there was some concern that the flow was igniting at the interface between the copper and stainless steel ducts, indicating that this junction may have been strongly influencing flame holding in the combustor. The plane 5 measurements indicate that the flow is auto-igniting upstream of the junction.

The plane 5 mole fraction plots show that, at this location, the fuel jet plume’s edge has penetrated nearly to the lower duct wall, as shown by the deficit in N$_2$ mole fraction near the bottom wall. In the initial analysis of this plane 5 data an unexpected bifurcation of the plume was observed in the O$_2$ and N$_2$ distributions. Upon closer inspection, this was caused by including in the data an incorrect $y$ value for one of the scans. By correcting this error in accordance with the original data logbook, the expected round plume was obtained, as seen in the figure.

At plane 6, some fuel products have penetrated to the bottom of the duct, distorting the plume near the duct floor. The minimum value of N$_2$ is higher than for the previous planes, as would be expected from a flow that is becoming better mixed as it flows along the duct. The region of reduced O$_2$ mole fraction corresponds to the region of highest temperature in the temperature map. This supports the view that combustion is strongest in that region of the flow.

Plane 7 looks more symmetrical left-to-right than planes 5 and 6, where the fuel jet plume is shifted slightly towards the right of the duct. The difference is particularly noticeable in the temperature distribution. The previously-mentioned repair to the nozzle required both the copper and steel sections of the combustor to be removed. Plane 7 properties were measured after the model was re-assembled, whereas planes 3, 5, and 6 (and the data of Ref. 1) were measured prior to this; it is likely that the duct sections were better aligned for plane 7 than for
the previous tests, resulting in a more symmetric distribution. This is consistent with the results of Ref 1 which were slightly asymmetrical at planes 5, 6, and 7.

**Table 1.** Statistical 95% confidence interval half width uncertainties (CIHW) in fitted surfaces, and results of the Model F-statistic (Mod F) and Lack of Fit P-statistic (LOF P) tests.

<table>
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<th>Plane 5</th>
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Table 1 summarizes the uncertainty in the fits for these mean quantities. These values represent the 95% CIHW uncertainty in the fit’s prediction of the mean surface, averaged over the entire surface of a given plane. For these mean property maps, the statistical uncertainties are typically nearly twice the systematic errors discovered during the flame and furnace calibration studies. For example, typical temperature measurement fit uncertainties were 25-50 K, whereas systematic errors were on the order of 30 K. Similarly, the statistical fit uncertainties for N₂ and O₂ mole fraction were between 0.003 and 0.017, whereas the systematic errors were less than or equal to 0.01. These results indicate that acquiring a factor of 2 or more data per plane (reducing these statistical uncertainties by a factor of 1.4) would have been a sensible investment of resources. However, acquiring more than 4 times more data would reduce the statistical errors to a level lower than the systematic errors, which would probably be a poor use of resources.

Table 1 also shows whether each fitted surface passed or failed the Model F-statistic and Lack of Fit P-statistic tests as described in References 28 and 29. Failure of the Model F-statistic test indicates poor signal-to-noise ratio in the surface being fitted. All 15 of the mean surface fits passed the Model F-statistic test, indicating good signal-to-noise in the fits. Failure to exceed a Lack of Fit (LOF) P-statistic of 0.05 indicates that a fit may contain significant lack of fit – that is, the response surface may be a poor fit to the data. Three of the 15 mean surfaces fitted failed this LOF P-statistic test. Truncating steep gradients at the edge of the data domain and refitting resulted in a passed LOF P-statistic. Alternately, choosing other functional forms produced slightly better fits that pass this test, in some cases. However this was not done because it was more consistent to use the same basis functions for all of the surfaces, to avoid introducing another variable into the analysis. Since these statistics depend on the overall quantity of data obtained, it is possible that taking a factor of 2 or more data would have resulted in fits that passed the LOF P-statistic test.

Spatial Variation in Fluctuating Parameters

As mentioned in the introduction, one of the main benefits of dual-pump CARS as a measurement technique is that it allows more than one flow property to be measured simultaneously. This capability allows us to determine statistical correlations between each of the measured flow properties in the duct. Such information is useful in the development of turbulence models for CFD simulations, and also provides important information about mixing in this particular combustor.
We classify the correlations into three categories: variances, such as $<T'T'>$,
$<\chi_O'\chi_O'>$ and $<\chi_N'\chi_N'>$, covariances between the various mole
fractions, such as $<\chi_N'\chi_O'>$, and covariances between the temperature and
various mole-fractions, such as $<T'\chi_N'>$ and $<T'\chi_O'>$. Surfaces of the square root of the variances and
covariances are presented in Figs. 8 and 9. Each of these plots has regions of low fluctuation at the edges of the
plot, where the gas is mostly freestream, and in the center of the plot, where much of the flow is H$_2$ from the fuel jet.
Between these two regions, all four maps show regions of large fluctuations, particularly along the sides of the fuel
plume. The shear between the plume and the freestream generates large fluctuations in both temperature and species
concentration.

Compare the plane 7 $T'T'$ map shown in Fig. 8(a) with the mean temperature map shown in Fig. 6. The
$T'T'$ map shows large temperature fluctuations near the bottom of the duct but much smaller fluctuations at the
top. The mean temperature distribution contains peaks at the top and bottom portions of the duct. This combination
indicates that turbulent mixing and combustion are still taking place at the bottom of the duct as evidenced by large
temperature fluctuations, but the hot flow at the top of the duct has become more evenly mixed and burnt. This
result is consistent with observations made in Ref. 1.

Consider the temperature/mole-fraction covariance maps, summarized in Fig. 9. At plane 3, the $T'\chi_N'$ and
$T'\chi_O'$ maps show strong positive correlations at the center of the fuel jet plume, meaning that high temperatures
correspond to high mole fraction of N₂ and O₂ and vice-versa. Recall that the fuel is injected at a temperature well below the freestream temperature, so that instantaneously low measured temperatures correspond to instantaneously high mole fraction of H₂ and low mole fraction of the remaining species. Elsewhere in plane 3, the correlations are approximately zero. For the most part, the \(<T'\chi_{O2}'>\) and \(<T'\chi_{N2}'>\) covariance maps are similar to each other. However for planes 6 and 7, the \(<T'\chi_{O2}'>\) covariance map shows some regions of significant negative correlation. This means that in these regions the instantaneously hotter-than-average gas has lower-than average O₂ content. Presumably, higher temperature coincides with more combustion, and more consumed O₂. The regions of large positive correlation at the bottom of the duct may, using the same argument, indicate that the instantaneous concentration of O₂ is controlled by the process of mixing with air rather than by combustion. Many of the insights drawn from these plots are common sense, but the value of the plots lies in knowing the magnitude and the location in the duct where these fluctuations occur.

Figure 9(c) contains the mole-fraction covariance maps for \(<\chi_{N2}'\chi_{O2}'>\). In planes 5 and 6, large positive correlations between N₂ and O₂ clearly identify the mixing layer around the fuel jet plume. However, at the center of the plume N₂ and O₂ are uncorrelated. This is sensible, since the bulk of O₂ that has diffused to the center of the fuel jet plume would have combusted.

Reference 29 showed that many more measurements are required to measure RMS fluctuations of properties with an acceptable uncertainty than are required to measure the means of these properties. The uncertainties of the
fits to these fluctuating parameters are also presented in Table 1, where they are once again averaged over the entire plane. On a percentage basis, CIHW values for these measurements varied from 10% to 15% of the maximum fitted values, averaged over the entire plane. Some of the fits fail the Model F and Lack-of-Fit P-Statistic tests. These tests could probably be passed if more data of the same quality was obtained (both statistics improve with larger data volume). The data is included as proof that such measurements can be made with the current system, although more measurements would be required to reduce the uncertainty to levels that would be acceptable for CFD model development and validation. In its present form the measurements of fluctuation in this data are sufficient to indicate the regions in the flow where interesting fluctuations in flow properties occur.

Table 1 also shows that just under half of the fluctuation maps shown in Figs. 8 and 9 pass the Model F-Statistic test, indicating that the signal-to-noise ratio in the data is insufficient to allow a good fit. Those that failed the fit only failed by a factor of two, indicating that, if twice as much data had been obtained, they probably would have passed the test. The fact that these fits failed the tests means that there are probably small amplitude variations shown in the graphs that are not real – rather they are fitting to noise. Nonetheless, the larger features shown in the maps are real. Furthermore, a few of the surface fits failed the Lack-of-fit P-Statistic test. As above, choosing different fit functions can result in satisfactory fits, but we did not do so, to avoid introducing another variable into the analysis.

**CONCLUSIONS**

We have used the dual-pump CARS measurement technique to simultaneously measure temperature and the absolute mole fractions of O\textsubscript{2} and N\textsubscript{2} in a model supersonic combustor. Nearly twelve thousand single-shot measurements were obtained in five different measurement planes oriented perpendicular to the flow direction. These measurements were fitted with analytic surfaces to provide two-dimensional maps of each mean flow property in each of the five planes. The maps were used, together with the raw data, to compute fluctuating quantities and their correlations, including $<T'\, T'>$, $<T'\, \chi_{N_2}'>$, and $<\chi_{O_2}'\, \chi_{N_2}'>$, among others. This type of data should be useful for developing new models to predict turbulent flow, mixing and combustion.

The standard deviation of the temperature measurement technique, as determined in flame and furnace calibration experiments, is less than the standard deviation of the freestream temperature, allowing an estimate of the
upper limit of facility flow temperature fluctuations to be made. These measurements indicate that standard deviations in the freestream temperature at the facility nozzle exit were ~130 K. Nozzle exit fluctuations in mole fraction were of the same order as the fluctuations measured in a premixed laminar flat flame, and were therefore not resolved by the technique. Upper limits for standard deviations of 0.024 and 0.042 were measured for \( \text{O}_2 \) and \( \text{N}_2 \) respectively.

The data presented here shows combustion being strongest at the top and bottom of the duct, spreading towards the sides of the duct as the flow proceeds downstream. Examination of the intensity of fluctuations about the mean allow us to determine where significant mixing and combustion are occurring in the flowfield.

The CARS system, analysis and application described above could be improved in several ways. First, the observed errors in \( \text{H}_2 \) mole fraction should be investigated so that the \( \text{H}_2 \) data obtained in the combustor can be analyzed and presented to complement the work outlined in the current paper. Measurements in a high-temperature static cell using several known \( \text{H}_2 \) concentrations at different temperatures would provide the necessary controlled environment for characterizing the systematic error. Second, the combination of an injection-seeded YAG laser and modeless dye laser may improve the precision of the measurements. Third, more data would reduce measurement uncertainty. In future work, an axisymmetric flowfield will be investigated, as the assumption of symmetry allows more precise determination of flow properties and their correlations with data sets of comparable size. Furthermore, a velocity measurement technique will be developed that operates simultaneously with the CARS method to provide much needed mean velocity profiles as well as correlations between velocities, temperatures and mole fractions.

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