



A TDLAS Sensor for Simultaneous Measurement of Temperature and C₂H₄ Concentration Using a Differential Absorption Scheme at High Temperature

Ting Zhang, Gang Zhang, Xin Liu, Guangzhen Gao and Tingdong Cai*

Jiangsu Key Laboratory of Advanced Laser Materials and Devices, College of Physics and Electronic Engineering, Jiangsu Normal University, Xuzhou, China

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*Correspondence:

Tingdong Cai
caitingdong@126.com

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Ethylene (C₂H₄) concentration and temperature is simultaneously measured in high temperature environments using a differential absorption scheme combined with a distributed feedback (DFB) diode laser near 1.620 μm. Based on optimal selection criteria, two wavelength pairs with absorption peaks at 6,174.58 cm⁻¹ and 6,174.98 cm⁻¹ are selected for the measurement. Temperatures are obtained from the ratio of the absorption cross-section of the two wavelength pairs, and C₂H₄ concentration is inferred from the differential absorption of one of the wavelength pairs. Measurements are performed in a high temperature cell ($T = 300\text{--}900\text{ K}$, $P = 1\text{ atm}$) to verify the accuracy of the system. The accuracies for the measurement of C₂H₄ concentration and temperature are 1.135 and 2.215%, respectively. Continuous time series measurements indicated that the system has a good stability, and the limit of detection achieved by Allan-Werle variance analysis is 6.6 ppm at the optimal average time of 340 s. All the measurements show the scheme is helpful for improving the practicality of laser absorption spectroscopy in combustion diagnosis.

Keywords: tunable diode laser absorption spectroscopy (TDLAS), differential absorption scheme, temperature, concentration, measurement

INTRODUCTION

Temperature is one of the scalar quantities on large technical combustion systems which directly reflects the combustion efficiency and affects the diffusivity of combustion products. The analysis of existing research [1–3] indicated that it is difficult to measure temperature or its distribution in combustion process. Therefore, it is necessary to develop an accurate and precise experiment approach to directly measure combustion temperature and provide time-resolved information of temperature distribution.

In combustion applications, planar laser-induced fluorescence (PLIF) is a kind of laser spectrum measurement technology for the diagnosis of high-temperature variable flow fields [4, 5]. PLIF technology can accurately measure the distribution of component concentration, temperature change, and other information in the combustion environment, but PLIF equipment is complex, expensive, and difficult to apply in industrial applications. Tunable diode laser absorption spectroscopy (TDLAS) is a valuable tool for studying the combustion environment due to its

non-contact measurement, rapid response time, and high sensitivity [6–9]. TDLAS technology has been demonstrated previously to provide quantitative measurements for species concentrations and temperatures in various combustion systems [8–11]. For example, Almodovar et al. identified the absorption transitions of nitric oxide (NO) on species sensing and two-color thermometry in high temperature applications, and some new insights to the temperature dependence of NO collision broadening were provided [12]. Sun et al. demonstrated the measurement of multi-species concentrations and temperature in a slagging coal gasifier with high pressure and high temperature using TDLAS, proving the capability of TDL sensors to monitor transient changes in gas temperature and the syngas heating value [13]. Cai et al. demonstrated accurate and fast measurement of carbon dioxide concentration and temperature based on TDLAS at high temperature and high pressure [14].

Through an investigation of relevant literature, it has been found that there are many studies on measurements of species concentration and temperature by direct absorption spectroscopy (DAS) or wavelength modulation spectroscopy (WMS) employing isolated absorption lines of the measured gas molecules [12–19]. However, for the molecules with dense absorption spectral features (such as ethylene and methane, which contain hydrocarbon bonds), there will be self-mixing or overlap with other molecules at an elevated temperature and pressure, making it difficult to accurately infer the complete transition properties through a single line or line pair. Therefore, the previous measurements of these species are mainly performed at low pressure or by employing a fixed-wavelength scheme. For example, Ren et al. performed fixed-wavelength measurements of ethylene at 10.532 μm using a CO₂ gas laser [20]. Strand et al. established a fixed-wavelength methodology for the measurement of a quantitative absorption cross-section of elevated temperature and pressure gases using an external-cavity quantum cascade laser with rapid-tuning mode and broad-scanning range [21]. Tanaka et al. constructed a sensor for trace gas analysis of C₂H₄ in combustion emission using WMS coupled with a DFB interband cascade laser near 3.356 μm under 3 kPa pressure [22]. The peak of the absorption may be shifted due to the change of the pressure in a practical combustion; these methods are difficult to implement for most sensors deployed in real-world environments.

In addition to the challenges of the conditions of the species being tested, it is also necessary to consider the interference caused by other species and soot in a practical combustion. To address this disadvantage, a differential absorption (or peak to valley) scheme is proposed to eliminate self-interference of the tested species or interference of other molecules with absorption features in the wavelength range of interest [23, 24]. And the method can also eliminate the effects of extinction or scattering caused by particles or liquid droplets. However, it has only been used for interference-free measurements of some species concentrations rather than temperature. In this study, we will demonstrate the simultaneous measurement of C₂H₄ concentration and temperature using TDLAS combined with the differential absorption scheme in an elevated temperature environment.

EXPERIMENTAL DETAIL

For high precision determination of temperature and C₂H₄ concentration, a schematic of the optical setup is shown in **Figure 1**. A continuous-wavelength DFB laser with a relatively narrow linewidth of about ~2 MHz operating at 1.620 μm is employed as a light source for this experiment. The laser is controlled by a low-noise laser driver (ILX Lightwave LDC-3724C) to probe the C₂H₄ absorption lines, and the laser temperature is fixed at 13.8°C while the current is scanned by a triangle voltage ramp generated by a function generator (RIGOL, DG1032Z). The laser beam is connected to a fiber beam-splitter to form two parts. The first part (about 50%) is propagated into a wavemeter (Bristol-671B-MIR) with a repeatability of ±1 ppm to record the frequency of the spectroscopic lines. The second part is collimated and introduced into a heat static cell to sense C₂H₄. The transmitted laser beams is directly recorded by a home-made detector. Finally, the output signal from the photodetector is acquired by a DAQ card (National instruments, USB6361).

The heat static cell is previously described in our previous work [25]. Briefly, the temperature of the cell can reach up to 1,200 K and pressure can withstand 10 atm under ideal conditions. The gas temperature in the static cell is determined by the average of three thermocouples with accuracies of 1% and these thermocouples are arranged equidistantly along the cell body. Before each experiment, the heat static cell is purged with pure N₂ for a few minutes to remove the residual gas. The pressure of the test gases is measured by a gauge with an accuracy of 0.25%. In this work, the cell is operated at 300–900 K at an operational pressure of 1 atm. When the temperature is stabilized, the C₂H₄-N₂ gas with different concentrations are delivered to the cell for measurement. Each spectrum is obtained with 30 average sweeps to increase the signal noise ratio (SNR).

THEORY AND METHODOLOGY

Algorithms for Data Processing

The theoretical details for concentration measurement with the differential absorption scheme were previously described in Ref. [26–28]. Here we will present a brief review and develop the theory for two-wavelength temperature measurement.

As a laser beam passes through a uniform gas mixture, the relationship between the transmitted intensity and incident intensity of the laser beam can be described using the Lambert Beer law:

$$-\ln\left(\frac{I}{I_0}\right)_v = \sigma(v, T, P) x \frac{P_{tot}}{RT} L = \alpha(v, T, P) \quad (1)$$

in which $\frac{I}{I_0}$ is the ratio of transmitted and incident laser intensities, $\sigma(v, T, P)$ (m² mol⁻¹) is the absorption cross section of target molecules, x is the mole fraction of the absorber, R (8.314 J K⁻¹ mol⁻¹) is the universal gas constant, P_{tot} (atm) and T (K) is the total gas pressure and the temperature, L (m) is the path length of the gas sample, and $\alpha(v, T, P)$ is the absorbance.

However, in certain situations, other absorbing species can produce absorption and interference absorption in the selected

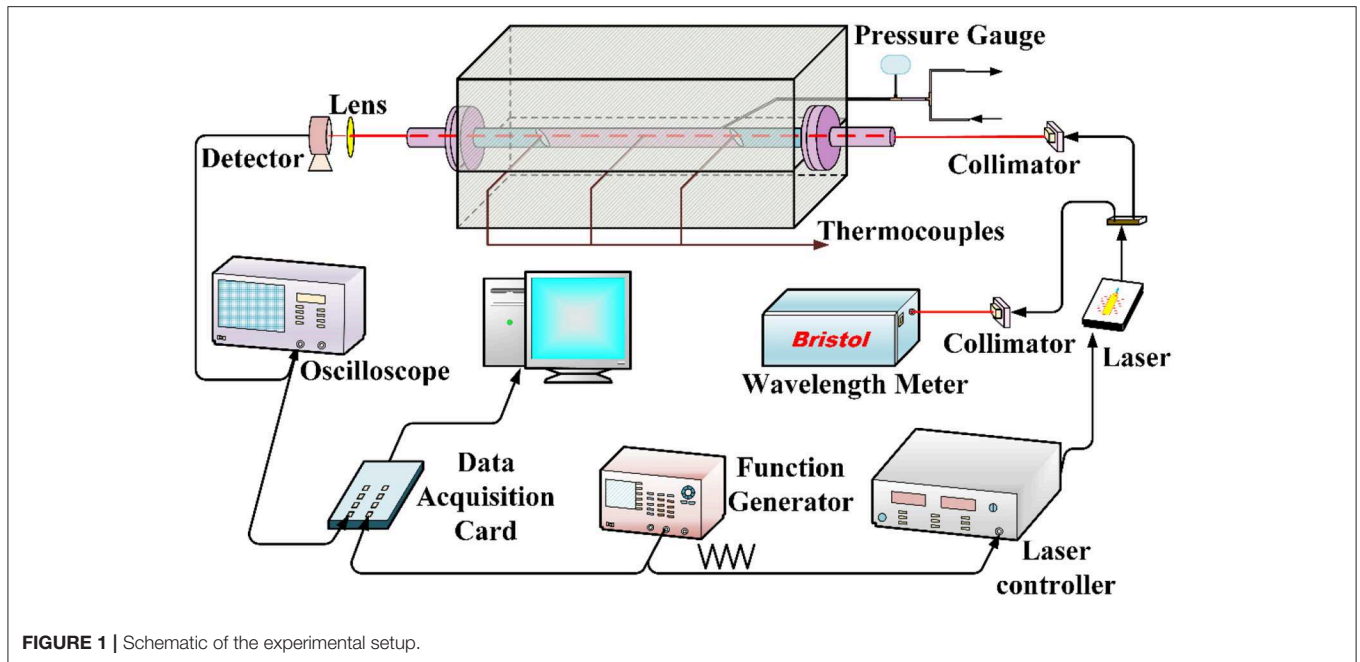


FIGURE 1 | Schematic of the experimental setup.

wavelength range, while extinction or scattering can also be caused by particles or liquid droplets. The Equation (1) can be modified to describe these effects:

$$-\ln\left(\frac{I}{I_0}\right)_v = \alpha_{target} + \alpha_{interference} + \tau_{extinction} \quad (2)$$

where α_{target} is the absorption by the target molecules, $\alpha_{interference}$ is the absorption from other interfering molecules, and $\tau_{extinction}$ includes the total extinction and scattering from all other sources. In the differential absorption scheme, the fractional transmissions should be acquired at the peak $(-\ln(\frac{I}{I_0})_{v,peak})$ and valley $(-\ln(\frac{I}{I_0})_{v,valley})$ wavelength.

In order to quantitatively measure the C₂H₄ concentration and temperature based on direct absorption spectroscopy, the absorption transitions with minimal interference from other species should be identified in the wavelength range of interest. The differential absorption $(-\ln(\frac{I}{I_0})_{v,peak}) - (-\ln(\frac{I}{I_0})_{v,valley})$, can be effectively applied to eliminate the influence of interference from other species and extinction while their magnitudes are almost the same at the peak and valley of the absorption feature of the measured molecule. Therefore, the mole fraction of the target substance can be calculated by subtracting the valley from the peak value of the absorption transition using Equation (2) at a specific temperature and pressure. After simplification, the following expression can be obtained:

$$x = \frac{-\ln\left(\left(\frac{I}{I_0}\right)_{v,peak} \left(\frac{I}{I_0}\right)_{v,valley}\right)}{\left(\sigma_{(v,peak)} - \sigma_{(v,valley)}\right) P_{tot} L / RT} \quad (3)$$

For a single absorbing species, the ratio of peak-to-valley absorbance is only a function of the differential absorption cross-section at the selected wavelength pairs.

$$\begin{aligned} \text{Ratio} &= \frac{-\ln\left(\frac{I}{I_0}\right)_{v_1,peak} - (-\ln\left(\frac{I}{I_0}\right)_{v_1,valley})}{-\ln\left(\frac{I}{I_0}\right)_{v_2,peak} - (-\ln\left(\frac{I}{I_0}\right)_{v_2,valley})} = \\ &= \frac{(\sigma_{v_1,peak} - \sigma_{v_1,valley})x n L}{(\sigma_{v_2,peak} - \sigma_{v_2,valley})x n L} = \frac{\sigma_{v_1,peak-valley}}{\sigma_{v_2,peak-valley}} \end{aligned} \quad (4)$$

where v_1 and v_2 represent the selected two wavelength pairs, respectively.

Since the absorption cross-section is strongly temperature-dependent [29], the cross-sections of peak and valley at selected wavelengths can be determined in a temperature range required in the work. The temperature dependences of the differential absorption cross-sections can be modeled with power law expressions based on the measured cross-section data.

$$\sigma_{v_1,peak-valley}(T) = \sigma_{v_1,peak-valley}(T_0) \left(\frac{T_0}{T}\right)^a \quad (5)$$

$$\sigma_{v_2,peak-valley}(T) = \sigma_{v_2,peak-valley}(T_0) \left(\frac{T_0}{T}\right)^b \quad (6)$$

where $\sigma_{v,peak-valley}(T_0)$ is the differential absorption cross-section at the reference temperature T_0 , a and b are the indices of the polynomial expression.

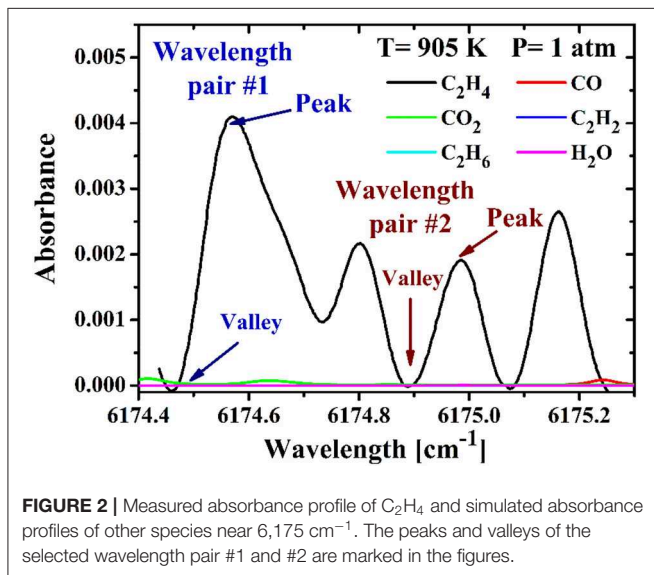
Since Equation (4) can be expressed as:

$$\text{Ratio} = \frac{\sigma_{v_1,peak-valley}}{\sigma_{v_2,peak-valley}} = \frac{\sigma_{v_1,peak-valley}(T_0)}{\sigma_{v_2,peak-valley}(T_0)} \left(\frac{T_0}{T}\right)^{a-b} \quad (7)$$

Finally, when the total pressure and the ratio are known, the gas temperature can be deduced.

Wavelength Pairs Selections Methodology

Selecting optimal absorption transitions is crucial to the performance of all TDLAS sensors. In this work, the main line selection criteria for the differential absorption-based system include strong absorbance at the peak wavelength, isolation from interference of the other species at the target range of interest wavelength range, adequate intervals for each wavelength pair, and the monotonicity between the ratio of the differential absorption cross-sections of the two wavelength pairs and temperature [24]. Here two wavelength pairs near 6,175 cm⁻¹ are selected based on those criteria. **Figure 2** shows the measured spectrum of C₂H₄ and the simulated absorbance of the main interference species in combustion based on HITRAN 2016 at 905 K and 1 atm, the selected wavelength pairs are marked in this figure. Since



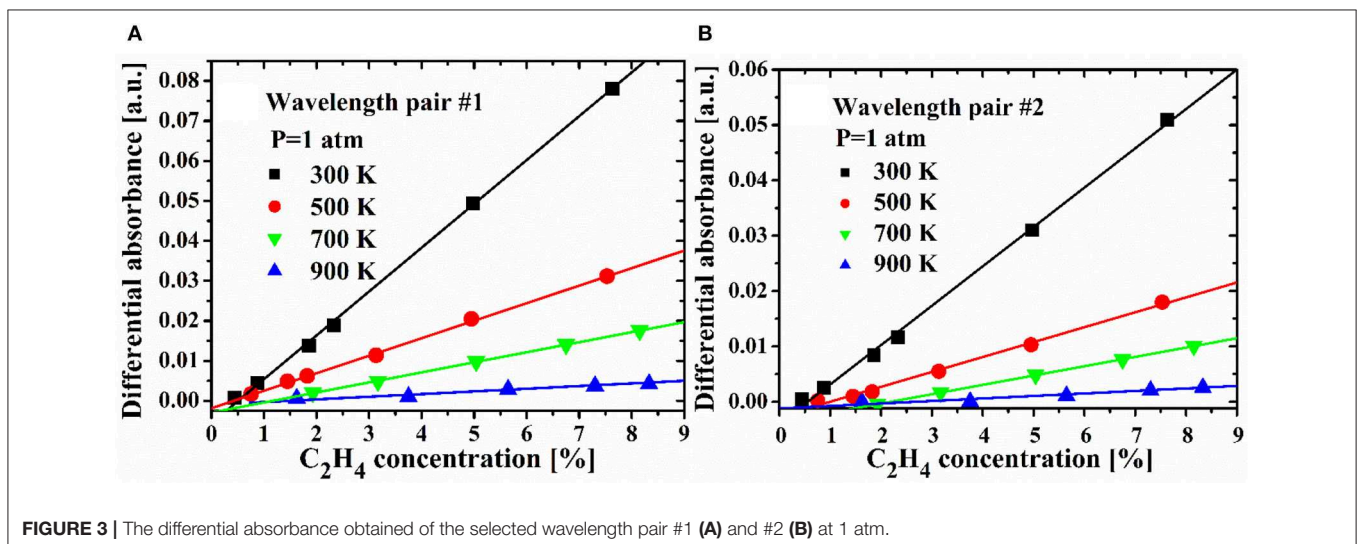
the absorbance of these interference species is almost the same at the marked locations, the interference absorption in combustion could be minimized using the differential absorption technique.

RESULTS AND DISCUSSION

Measurement of Differential Absorption Cross Sections

In order to obtain the differential absorption cross-sections for the selected two wavelength pairs, direct absorption spectra of a set of C₂H₄/N₂ mixture with different concentrations are recorded in the temperature range of 300–900 K at normal atmosphere. For each wavelength pair, the differential absorption cross-sections can be inferred from the differential absorbance between the peak and valley. The differential absorbance of wavelength pair #1 and #2 vs. the C₂H₄ concentration is plotted in **Figure 3**. According to Equation (3), the absorption cross-section of each wavelength pair can be determined from the linear fit to the measured differential absorbance at various C₂H₄ concentrations. It also can be seen from the figure that the differential absorbance linearly increases with the C₂H₄ concentration, but it decreases with temperature at a certain C₂H₄ concentration. As the temperature increases, the temperature dependence increases. This is because the transitions overlap each other as the temperature increases, and the profile of the spectrum becomes relatively dense at high temperatures.

Figure 4 shows the absorption cross-sections at peaks and valleys, and differential cross-sections for the used C₂H₄ at selected wavelength pairs, which obtained at different temperatures and atmospheric pressure. As indicated in **Figure 4**, the differential cross-section ($\sigma_{\text{peak-valley}}$) of the selected two wavelength pairs show strong temperature dependence. Both of them decrease with temperature. Then the generalized expression for differential cross-sections as a function of temperature can be



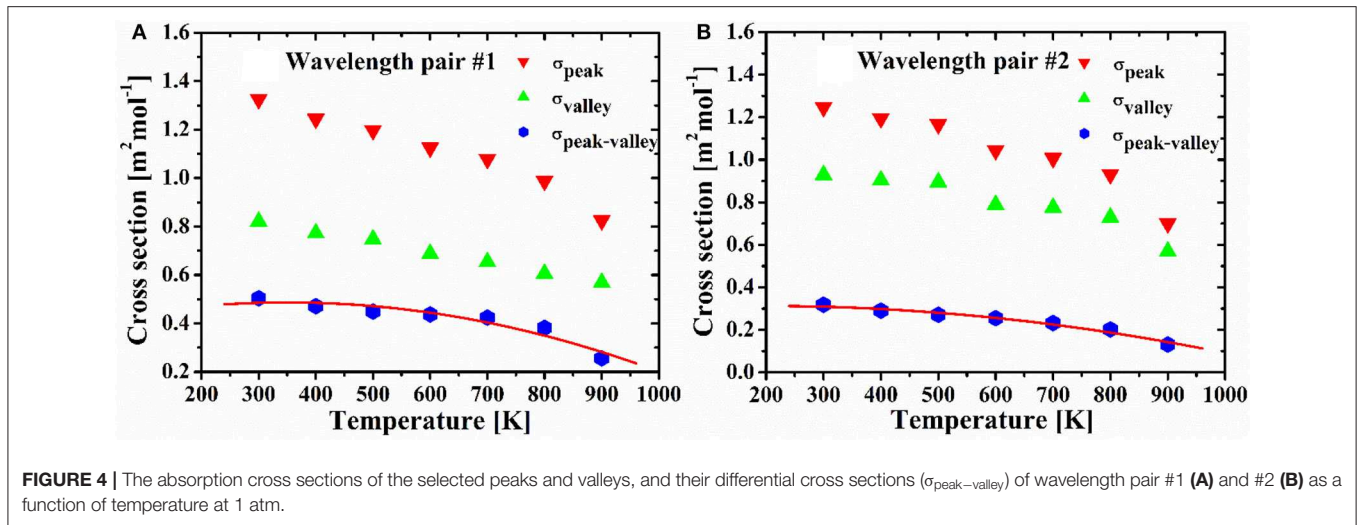


FIGURE 4 | The absorption cross sections of the selected peaks and valleys, and their differential cross sections ($\sigma_{\text{peak-valley}}$) of wavelength pair #1 (A) and #2 (B) as a function of temperature at 1 atm.

derived for the two selected wavelength pairs.

$$\text{For wavelength pair \#1: } \sigma_{\#1}(T) = \sigma_0 \left(\frac{T_0}{T} \right)^{0.386},$$

$$\text{where } T_0 = 305 \text{ K}, \sigma_0 = 0.528 \text{ m}^2 \text{ mol}^{-1}.$$

$$\text{For wavelength pair \#2: } \sigma_{\#2}(T) = \sigma_0 \left(\frac{T_0}{T} \right)^{0.544},$$

$$\text{where } T_0 = 305 \text{ K}, \sigma_0 = 0.336 \text{ m}^2 \text{ mol}^{-1}.$$

The differential absorption cross sections derived from the expressions agree well with the measured values, and the standard deviations between the derived and measured values are 4.67% for pair #1 and 2.89% for pair #2.

Measurement of Temperature and C₂H₄ Concentration

Experiments with a controlled C₂H₄-N₂ mixture over the measured range of temperature are performed to validate the accuracy of temperature and C₂H₄ concentration, inferred from the differential fractional transmission. The top row of **Figure 5** shows the comparison between the measured temperatures and the average values recorded by three thermocouples. Each data point represents the mean of temperature obtained from multiple measurements. It can be seen that the measured temperatures agree well with the reading of the thermocouple in the experimental temperature range.

The consistency of measurement resulting at low temperature is better than that at high temperature, which can be explained by the reduction of SNR at high temperature. Correlation of the measured data points has an R-squared value of 0.999, and the mean value of the standard deviations is 1.135%. The linear fitting slope is 0.997 ± 0.002 , and there is an average bias about 4 K between the measured temperature and the thermocouple reading. The bottom row of **Figure 5** shows the ratio of the measured C₂H₄ concentration (X_{measured}) and the recorded C₂H₄ concentration during gas premixing ($X_{\text{referenced}}$). The C₂H₄ concentrations are calculated using the wavelength pair #1

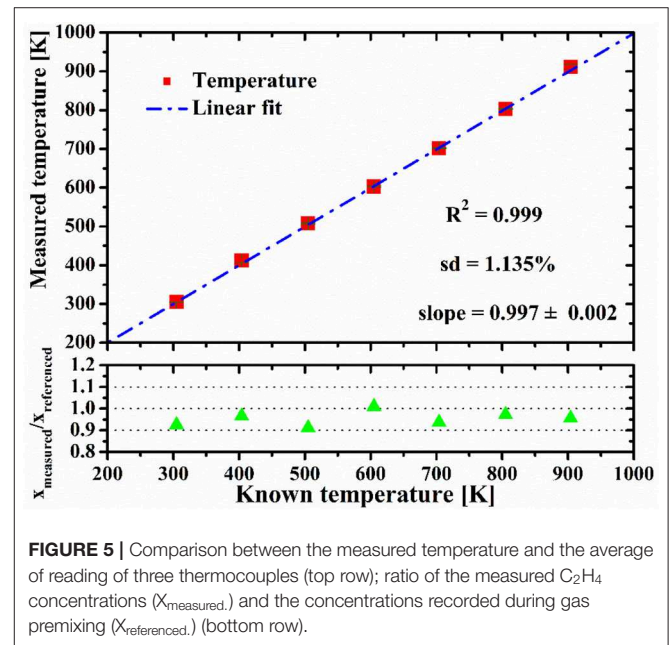
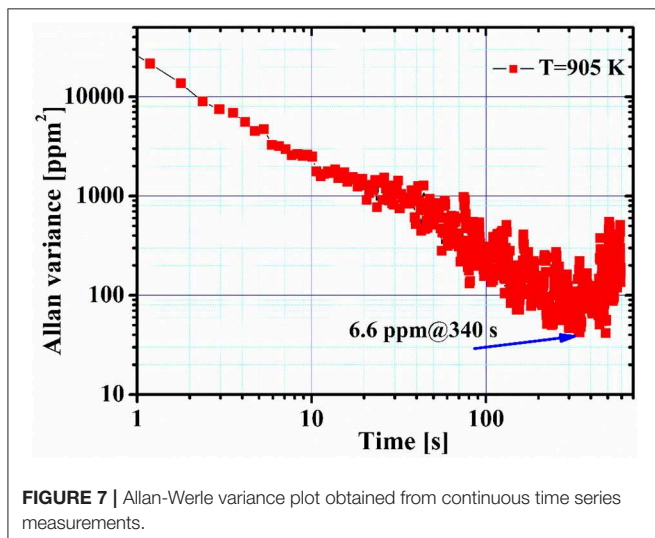
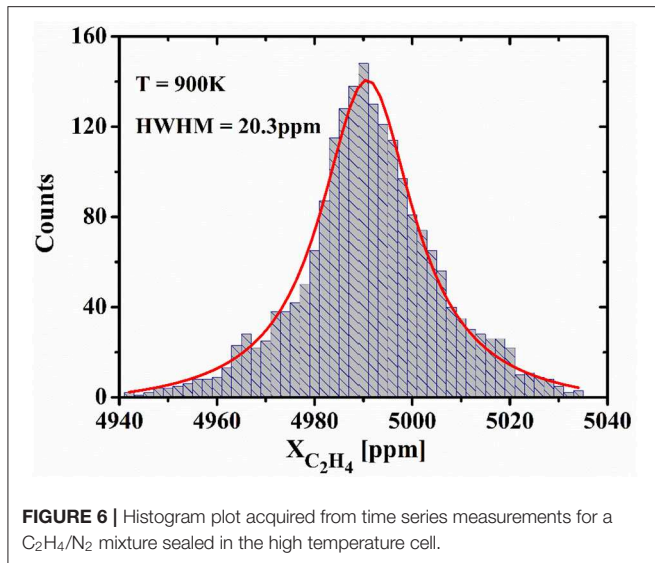


FIGURE 5 | Comparison between the measured temperature and the average of reading of three thermocouples (top row); ratio of the measured C₂H₄ concentrations (X_{measured}) and the concentrations recorded during gas premixing ($X_{\text{referenced}}$) (bottom row).

which have bigger differential absorption. The average standard deviation is 2.215% between the measured and reference values of C₂H₄ concentration. The reasonable consistency between the measured and referenced values confirms the practicability of the differential absorption technique for measurement of temperature and C₂H₄ concentration in this work.

Precision and Sensitivity

In order to validate measurement precision and stability, time series measurement is performed for a C₂H₄/N₂ mixture with concentration of 0.5% sealed in a high temperature cell during 33 min. The measured data was obtained for each 1 s, and 2,000 data points are obtained in the measurement.



The histogram plot of measured data used to evaluate the measurement precision for C₂H₄ concentration is shown in **Figure 6**. As shown in this figure, the histogram demonstrated a good Gaussian distribution, and the measurement precision for C₂H₄ concentration is 20.3 ppm, which can be determined by the half width at half maximum (HWHM) of the Gaussian profile [15].

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The Allan-Werle analysis can be used to evaluate the sensitivity of the system [30]. **Figure 7** shows the Allan-Werle variance obtained from the successive measurement to validate the sensitivity of the measurement. The Allan-Werle analysis shows when the average time of the system is 340 s, the corresponding minimum detection limit of the C₂H₄ concentration can be down to 6.6 ppm. The high sensitivity and measurement precision confirm the good performance of the system based on differential absorption scheme.

CONCLUSION

A TDLAS based system for simultaneous measurements of C₂H₄ concentration and temperature is achieved with the help of the differential absorption scheme which can eliminate the inferences from other species. The measurements are performed with C₂H₄/N₂ mixture of different concentrations in a static cell from 300 to 900 K at normal atmosphere. The comparison between the measured values and referenced values confirms the accuracy and reliability of the system. The measurement precision for C₂H₄ concentration is 20.3 ppm at 900 K. According to the Allan-Werle variance analysis, a sensitivity of 6.6 ppm can be achieved with an averaging time of 340 s. In the future, we will improve the performance of the system to extend its applicability to study the fields of the shock tube, the internal combustion engine, or flames of different types.

DATA AVAILABILITY STATEMENT

The datasets generated for this study are available on request to the corresponding author.

AUTHOR CONTRIBUTIONS

TZ was responsible for the experimental setup and writing. GZ and XL were responsible for the data processing and picture editing of this paper. GG was responsible for the verification of the overall experiment, and TC was responsible for the revision of the paper. All the authors made positive contributions to the work.

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Conflict of Interest: The authors declare that the research was conducted in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest.

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