Influence of light coupling configuration and alignment on the stability of HWG-based gas sensor system for real-time detection of exhaled carbon dioxide

Tao Zhou, Tao Wu*, Huailin Zhang, Qiang Wu**, Weidong Chen, Chenwen Ye, and Xingdao He

Abstract—A mid-infrared tunable diode laser absorption spectroscopy (TDLAS) gas sensor based on hollow waveguide (HWG) gas cell for real-time exhaled carbon dioxide (eCO2) detection is reported. A 2.73 μm distributed feedback (DFB) laser was used to target a strong CO2 absorption line, and wavelength modulation spectroscopy (WMS) with the second harmonic (WMS-2f) was used to retrieve the CO2 concentration with high sensitivity. The influence of different parameters, including coupling configuration of HWG, laser-to-HWG and HWG-to-detector coupling alignment on the stability of the HWG sensor is systematically studied. The HWG eCO2 sensor possesses low sensitivity and faces interference from water vapor and hydrocarbons. Beyond that method, high finesse optical cavity and multi-pass cell significantly increases the absorption path length and improves the detection sensitivity. However, these conventional systems have disadvantages of rigid requirement of precision optical light alignment, high cost, large physical size and especially large volume (usually larger than 100 ml) resulting in unsuitable for real time exhaled gas measurement. Laser absorption spectroscopy has been widely used in gas detection and plays an important role in industrial process analysis, environmental pollution monitoring, scientific research and other fields. Due to its medical interest, in recent years a variety of spectroscopy techniques have been developed to detect CO2 and 13CO2/12CO2 isotopic ratio in exhaled breath. Non-dispersive infrared spectroscopy (NDIR) is commonly used for the detection of eCO2 [5]-[7]. NDIR measures the gas concentrations by detecting the infrared light attenuation using a photo detector. Commercial NDIR CO2 capnographs work very well with high target gas selectivity and do not need frequent calibration, however, it usually possesses low sensitivity and faces interference from water vapor and hydrocarbons. Beyond that method, high finesse optical cavity based absorption spectroscopy, such as cavity ring-down spectroscopy (CRDS) [8] and integrated cavity output spectroscopy (ICOS) [9], and tunable diode laser absorption spectroscopy (TDLAS) with a multi-pass cell have also been widely used for eCO2 detection [10], [11]. The use of high finesse optical cavity and multi-pass cell significantly increases the absorption path length and improves the detection sensitivity. However, these conventional systems have disadvantages of rigid requirement of precision optical light alignment, high cost, large physical size and especially large volume (usually larger than 100 ml) resulting in unsuitable for real time exhaled gas measurement [9], [12]. Hollow waveguide (HWG) possesses a low volume and an extended optical path length that gives absorption spectroscopy a fast response time and high sensitivity with lower cost [13], [14]. Several authors have reported the use of HWG gas cells in absorption spectroscopy using TDLAS or Fourier transform infrared spectroscopy (FTIR). Xiong et al. [15] designed an HWG based CO2 sensor for capnography using direct absorption spectroscopy. Kim et al. [16]
established a gas sensor with an HWG combined with an FTIR spectrometer for CO2 and CH4 detection. All present studies are mainly focused on the improving of the laser to HWG coupling efficiency of laser to HWG [17], [18]. However, stability is a significant parameter for an HWG-based sensor system, which is mainly limited by temperature drifts, mechanical vibrations, and moving fringes in background spectra, and usually described by the Allan variance. However, the investigation of the effect of laser-to-HWG and HWG-to-detector coupling alignment on stability of the HWG-based sensor system is missing in the previous literatures. In this paper, a mid-infrared HWG eCO2 sensor employing a 1 m long HWG was developed for real-time eCO2 detection. Second harmonic (WMS-2f) wavelength modulation spectroscopy (WMS) was applied for eCO2 detection. A systematic study of the influence of sensor system parameters, including coupling configuration of HWG, laser-to-HWG and HWG-to-detector coupling alignment on the stability of the HWG sensor was performed. Our study shows that, Laser-to-HWG and HWG-to-detector coupling alignment play more important role to the stability of the sensor system, which provides a new guide for improving stability of the system. Finally, real time experiment for eCO2 was conducted to evaluate the performance of HWG eCO2 sensor for medical requirement.

II. WMS METHOD

The application of WMS in TDLAS can effectively improve the detection sensitivity and has been widely used in the measurement of absorption spectroscopy [19]. In WMS-2f method, the injection current of the DFB laser is modulated by the sawtooth wave superimposed with a high-frequency sinusoidal signal. The frequency (wavelength) ν(t) and intensity I₀(t) of the laser are also modulated accordingly. Their Fourier expansions are given by

\[ \nu(t) = \bar{\nu} + a \cos(2\pi ft) \quad (1) \]

\[ I_0(t) = \bar{I}_0 \left[ 1 + i_0 \cos(2\pi ft + \psi_0) + i_2 \cos(2 \cdot 2\pi ft + \psi_2) \right] \quad (2) \]

Where \( \bar{\nu} \) is the average laser frequency (wavelength); \( a \) and \( f \) are the amplitude of wavelength modulation and modulation frequency, respectively; \( \bar{I}_0 \) is the average laser intensity; \( i_0 \) and \( i_2 \) are the Fourier coefficients of the linear and non-linear intensity modulations; \( \psi_0 \) and \( \psi_2 \) are the phase shift of the linear and non-linear intensity modulations, respectively. For weak absorption (\( \alpha(\nu) < 0.05 \)), the absorption can be expanded into the Fourier cosine.

\[ \alpha(\nu + a \cos(2\pi ft)) = -\sum_{k=0}^{\infty} H_k(\nu, a) \cos(k \cdot 2\pi ft) \quad (3) \]

Here, \( H_k \) is the kth harmonic component of the absorbance \( \alpha \).

\[ H_k(\nu, a) = -\frac{1}{2\pi} \int_{-\pi}^{\pi} \alpha(\nu + a \cos \theta) \cos k\theta d\theta, k \geq 1 \quad (5) \]

For the WMS-2f detection, a reference signal with a frequency of 2f is sent to the lock-in amplifier, and the X-channel of the modulation signal (the in-phase signal) can be expressed as follows.

\[ X_{2f} = \frac{G I_{lo}}{2} \left[ H_z + \frac{i_0}{2} (H_z + H_i) \cos \psi_1 \right. \\
+ \left. i_2 \left( 1 + H_0 + \frac{H_i}{2} \cos \psi_2 \right) \right] \quad (6) \]

Where \( G \) account for the optical-electrical gain of the detection system. Usually, \( i_2 \) is much smaller than \( i_0 \). For an isolated transition, the odd term of the Fourier coefficient at the center of the spectrum line is zero. The relationship between the peak value \( X_{2f}(\nu) \) of the WMS-2f signal and the mole fraction \( x \) of the target gas can be calculated as.

\[ X_{2f}(\nu) \approx \frac{G I_{lo}}{2} H_z(\nu) \]

\[ = -\frac{G I_{lo} P S x L}{2\pi} \int_{-\pi}^{\pi} \phi(\nu + a \cos \theta) \cdot \cos 2\theta \cdot d\theta \]

Where \( P \) and \( L \) are the total gas pressure and the effective optical path length, respectively. \( S \) and \( \phi \) are the line strength and line shape function, respectively.

III. LINE SELECTION

According to the HITRAN database [21], CO2 has strong absorption lines in the mid-infrared region. For the eCO2 detection, the main interfering gas is H2O. Apart from H2O, the concentrations of other potential interference in breathing gases are \( 10^4-10^{10} \) times smaller than that of eCO2, which make their influence negligible. To eliminate the interference from H2O, the CO2 absorption line was selected at 3661.637 cm\(^{-1}\) with a line strength of 7.135 \( \times \) 10\(^{-22} \) cm\(^{-1}\)/(molecule cm\(^{-2}\)), which has no overlap with that of H2O. Fig. 1 shows the simulation of the absorption spectrum of 5% H2O and 5% CO2 for wavenumber range from 3657.57 cm\(^{-1}\) to 3665.95 cm\(^{-1}\) at a pressure of 270 torr and a temperature of 296 K. The inset of Fig. 1 shows the selected absorption line at 3661.637 cm\(^{-1}\). It is worth noting that the choice of absorption line also considers the characteristics of the laser. The stronger absorption line at 3663.851cm\(^{-1}\) has not been selected, because it is out of the wavelength tuning range of the laser used in our experiments.
IV. EXPERIMENT SETUP

The eCO2 sensor based on HWG is shown in Fig. 2(a). A DFB laser (Nanoplus GmbH, Germany) with a center wavelength of 2.73 μm and a maximum output power of 11.2 mW was used as an optical source. The laser was packaged in a TO-5 can with integrated Peltier and temperature sensor, which attached to an aluminum heatsink to remove heat dissipated from Peltier and the laser. A laser controller (ILX Lightwave, LDC-3724C, USA) was used to control the current and temperature of the laser diode. The current of the laser was set to 148.39 mA, and the operating temperature of the laser was set and controlled at 30 ºC. High-frequency sine wave (1.7 kHz) generated by a lock-in amplifier (Stanford Research System, SR830, USA) and a sawtooth wave (2 Hz) generated by a function generator were superimposed by an adder, and then sent to the modulation current port of the laser controller for performing WMS. The laser beam was collimated by a collimating lens and coupled through an aperture diaphragm into a 1 m long and 1 mm inner diameter HWG (Polymicro Technologies, HWEA10001600, USA) gas cell with the volume of 0.78 cm³, which was much smaller than that of the conventional multi-pass gas cell. The HWG fiber was firstly inserted into a fiber ferrule, and then inserted into the slot in the gas chamber and sealed with a layer of silicone rubber outside the chamber as shown in Fig. 2(b). The light output from the HWG cell was detected by a photodetector (VIGO System S.A., PVI-4TE-10.6, USA). The lock-in amplifier demodulated the output signal of the photodetector to obtain the WMS-2f signals, which were then acquired by a data acquisition card (ADlink, DAQ-2010, China) and further processed by a self-developed LabWindows program. The modulation parameters, such as modulation amplitude, modulation frequency, reference phase, time constant of lock-in amplifier, and laser scanning frequency, and gas pressure in the HWG were optimized by obtaining maximum lock-in amplifier, and laser scanning frequency, and gas pressure in the HWG were optimized by obtaining maximum modulation parameters, such as modulation amplitude, 50 mm. In the direct coupling configuration, the aperture size of the aperture diaphragm was sequentially set to 0.7 mm, 1 mm, 1.5 mm, 2 mm, 2.3 mm, where 0.7 mm is the minimum aperture that can be set by the aperture diaphragm used in our experiment, and 2.3 mm is the actual size of the laser beam.

V. THE COUPLING PATTERN OF LASER BEAM TO HWG AND ITS EFFECT ON SYSTEM SNR AND STABILITY

The coupling configuration between the laser beam and the HWG can be achieved in a focusing coupling configuration or in a direct coupling configuration (incident directly into the HWG). Here, the focusing coupling configuration and the direct coupling configuration are both studied. The influence of the laser spot size (modulated by an aperture diaphragm) on direct coupling configuration is explored. The relation between stability time and SNR of direct coupling configuration is investigated. The stability of the system for these two coupling mode is explored. The relation between stability time and SNR of direct coupling configuration is investigated.

5% CO2 was continuously pumped into the HWG cell, and the pressure in the cell was controlled at 270 torr. In the focusing coupling configuration, the laser beam was focused into the center of the HWG inlet by a lens with a focal length of 50 mm. In the direct coupling configuration, the aperture size of the aperture diaphragm was sequentially set to 0.7 mm, 1 mm, 1.5 mm, 2 mm, 2.3 mm, where 0.7 mm is the minimum aperture that can be set by the aperture diaphragm used in our experiment, and 2.3 mm is the actual size of the laser beam.
The WMS-2ƒ signals for focusing coupling and direct coupling with different spot sizes were recorded and shown in Fig. 3. It can be seen that the SNR for focusing coupling is highest. For direct coupling configuration, as the spot size decreases, the SNR decreases, this is due to a gradual reduction of the laser power. Fig. 4 shows the Allan variance obtained by measuring WMS-2ƒ signals. It can be clearly seen that, although the focusing coupling has highest SNR, the system stability for focusing coupling is worse (with higher variance limit) than that of direct coupling with 1 mm spot. As the spot diameter decreases from 2.3mm to 0.7mm, the system stability time for direct coupling is increased from 6 s to 57 s, which is possibly due to the fact that smaller aperture and acceptance angle collects center of the laser beam which has better laser beam quality illuminated to the HWG, resulting in small fiber model noise. However, the smaller aperture results in smaller SNR (as shown in Fig. 5), which is not conducive to improving the detection sensitivity (As the aperture is gradually reduced, the detection sensitivity is reduced from 7.9×10⁻⁶ cm⁻¹Hz⁻¹/² to 1.8×10⁻⁶ cm⁻¹Hz⁻¹/²). Here, direct coupling with the aperture of 1 mm reaches a compromise between good SNR and long stability time.

For direct outcoupling, possible parallel optical surfaces interference caused by focusing lens is avoided. To study the effect of partial collection of light from the fiber on system stability, the distance between the output of HWG and the detector was varied between -200μm and 200μm based on initial distance of 4cm. An optimal detector displacement distance Δz was found at -40μm (Fig. 6). It is noted that this optimal value obtained in the experiments is for our setup only. The detector is suggested to be placed as close as possible to the fiber end to collect all the light coming from the fiber end for any HWG-based setup. The influence of incoupling (position of the laser) on system stability was also investigated. The lateral displacement Δr between the center of laser beam and the center axis of HWG was varied from 0 to 120μm in 40μm steps. We found the laser at the central position given longest stability time (Fig. 7).
VI. INVESTIGATION OF RESPONSE TIME AND CONCENTRATION CALIBRATION

The response time of the HWG eCO2 sensor was investigated as shown in Fig. 8. Two mass flow controllers controlled the flow rate of pure CO2 gas (CO2 ≥ 99.999%) and pure nitrogen gas (N2 ≥ 99.999%) respectively. Five different samples of CO2 with concentrations varying from 0 to 6.3% were prepared by changing the flow rates of two mass flow controllers while maintaining a constant gas flow rate of 6.7 ml/s through the HWG cell. The pressure in the HWG cell was controlled to be 270 torr for obtaining WMS-2f signal. In the beginning, the HWG cell was full of pure nitrogen. Then, at 40 s, the diluted CO2 gas samples were filled into the HWG cell. The duration time of each concentration level gas sample was approximately 30 s to ensure that the measured signal was in a stable state. At the end of the measurement, pure nitrogen was again filled into the HWG cell. There was no significant drift found during the measurement period by the nitrogen signal levels. As shown in the insert of Fig. 8, the response time τr (10 - 90%) of the concentration levels in rising and falling process was about 2.7 s and 2.2 s, respectively, while the delay time τd (0 - 10%) was approximately in the same time of 0.6 s [22]. Due to the smaller volume of the HWG than the traditional gas cell (τr ~ 5 min) [12], the response time τr is much faster compared to the traditional gas cell with the similar gas flow rate. It is worth noting that the response time τr here also included the time for gas mixing time in the HWG cell, the actual response time of the HWG sensor was less than 2.7 s.

In order to obtain the relationship between WMS-2f peak values and CO2 concentrations, CO2 samples were prepared by using pure CO2 gas and 3000 ppmv standard CO2 gas diluted with pure N2, and the continuous measurements for different CO2 concentration samples were performed and averaged 5 times with a 100 ms acquisition time. Fig. 9(a) shows the WMS-2f signals at different CO2 concentrations with a 0.5 s temporal resolution. A linear relationship between WMS-2f peak values and CO2 concentrations was observed as shown in Fig. 9(b). Fig. 9(b) shows the fit curve have a good linearity for CO2 concentrations varied from 0 - 6.3%, even down to 200ppmv, which were used as calibration model to determine CO2 concentrations. The error bars of the WMS-2f peak values were in the order of 10^-3, which cannot be clearly seen in Fig. 9(b).
The measurement precision of the HWG eCO2 sensor can be analyzed by Gaussian distribution [23]. 300ppmv diluted CO2 gas sample was used as reference gas and pumped into the HWG cell. The gas pressure and flow rate were maintained at 270 torr and 6.7 ml/s, respectively. 600 measured data points were successively recorded and averaged 5 times corresponding to 0.54 s temporal resolution, as shown in Fig. 10(a). A histogram plot of CO2 concentration deviation which is fitted by a Gaussian profile is shown in Fig. 10(b). The measurement precision can be determined by the half width at half maximum (HWHM) of the Gaussian profile. It indicates that the measurement precision was 20.9 ppmv with 0.54 s temporal resolution. The corresponding average concentration value was 297 ± 20.9 ppmv. The measurement precision was mainly limited by random noise and etalon fringes superimposed on the WMS-2f signals. In addition, the small jitter of the HWG would lead to instability of the WMS-2f signals, which also affected the measurement precision.

VIII. CARBON DIOXIDE IN EXHALED BREATH

The eCO2 was measured in real time from one healthy volunteer in our lab and the results recorded with 0.54 s acquisition time are shown in Fig. 11. The pressure in the HWG cell was controlled to be 270 torr, and the breathing gas was introduced into the HWG cell by breath tube according to the direction of gas passage shown in Fig. 2. During the inhalation, the environment air was pumped into HWG through the side-port of breath tube, and the CO2 concentrations are stable and fluctuated around 360ppmv as shown in Fig. 11, hence the influence of ambient air on eCO2 detection can be negligible. In addition, the trends and values of concentration which are not affected by the breath rate changes were consistent with those in the literatures [24], [25]. It is noted that the response time $\tau$ of 3.7 s in this case is larger than that of 2.7 s described above. This is possibly due to the contribution of residual eCO2 in long breath tube during exhalation. If the volume of breath tube is made smaller and the length of breath tube is shorter, the response time will be faster.

IX. DETECTION LIMIT

The signal averaging method is a simple but practical method for eliminating background white noise. If a signal is averaged N times, the white noise may be reduced by a factor of $N^{1/2}$, which can be estimated by Allan variance [26]. Allan variance can evaluate the stability of the measurement system as well as the detection limit. Allan deviation was performed on a constant concentration of 300ppmv diluted CO2 sample which was pumped into HWG cell continuously, and Allan deviation (square root of variance) plot was shown in Fig. 12. The results shows that the Allan deviation decreases with the square root of the averaging time (shown in Fig. 12 as the red line of gradient = −0.5). The detection sensitivity was 17 ppmv with acquisition time of 0.54 s, corresponding to a detection sensitivity of $6.9 \times 10^{-8} \text{ cm}^2 \text{ Hz}^{-1/2}$, which indicates that the TDLAS sensor is applicable for clinical eCO2 monitoring. The minimum in the Allan deviation plot at 26 s represents the optimum integration time. The detection limit can reach as low as 1.7 ppmv, corresponding to a detection sensitivity of $1.3 \times 10^{-8} \text{ cm}^2 \text{ Hz}^{-1/2}$, as the integration time increases to 26 s. Thereafter the Allan deviation moves into a drift dominated region where it starts to increase with increasing averaging time. Furthermore, the detection limit can be further improved by increasing HWG length, reducing hardware noise and fixing the HWG firmly to suppress the etalon signal. Table 1 shows the detection limit and other experimental parameters.
obtained by using different methods to measure eCO₂. Though direct absorption spectroscopy (DAS) used in ref. [10], [11], [15] is calibration-free, the DAS is mainly limited by 1/f noise. The WMS uses a higher modulation frequency (~kHz) which suppresses the 1/f noise effectively, which is verified in Table 1 that the absorbance sensitivity (independent of optical length and related to stability of system) for WMA is usually higher than that for DAS. It is noted that the absorption sensitivity of the HWG sensor can be improved significantly by adding two high reflectivity mirrors to both ends of the HWG to form a high finesse resonant cavity, which effectively increases the length of optical path [8].

![Allan deviation plot for measured 300ppmv CO₂ sample.](image)

**Fig. 12.** Allan deviation plot for measured 300ppmv CO₂ sample.

**TABLE I**

<table>
<thead>
<tr>
<th>Reference</th>
<th>Wavelength (μm)</th>
<th>Pressure (torr)</th>
<th>Optical length (m)</th>
<th>Method</th>
<th>Detection limit/Absorbance sensitivity</th>
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<tbody>
<tr>
<td>[10]</td>
<td>4.91</td>
<td>380</td>
<td>26</td>
<td>TDLAS/NDIR</td>
<td>0.5%/~10⁻²</td>
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<td></td>
<td></td>
<td></td>
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<td>TDLAS (Multipass cell)</td>
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<td>[11]</td>
<td>2</td>
<td>760</td>
<td>0.025</td>
<td>TDLAS/NDIR</td>
<td>&lt;300 ppmv/~10⁻⁴</td>
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<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>TDLAS (Single cell)</td>
<td>&lt;300 ppmv/~10⁻⁴</td>
</tr>
<tr>
<td>[15]</td>
<td>2.003</td>
<td>760</td>
<td>0.12</td>
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<td></td>
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<td></td>
<td>TDLAS (HWG cell)</td>
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</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>TDLAS/NDIR</td>
<td>10 ppmv/~10⁻⁸</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>WMS</td>
<td>100 ppmv/~10⁻⁴</td>
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<td>[27]</td>
<td>1.575</td>
<td>150</td>
<td>40</td>
<td>TDLAS/NDIR</td>
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<td>WMS (Multipass cell)</td>
<td>100 ppmv/~10⁻⁴</td>
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<tr>
<td>[28]</td>
<td>4.7</td>
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<td>[7]</td>
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<td>[8]</td>
<td>1.6</td>
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<td>WMS (HWG cell)</td>
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<tr>
<td>Our present work</td>
<td>2.73</td>
<td>270</td>
<td>1</td>
<td>WMS/NDIR</td>
<td>1.7 ppmv/~10⁻⁶</td>
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**REFERENCES**


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