A Sensitive and Reliable Carbon Monoxide Monitor based on the Optimized Direct Absorption Spectroscopy Approach by using a 2.33 μm Laser Diode

Zhaowei Wang, Yanfang Li, Tingting Zhang, Jie Hu, Yin Wang, Yubin Wei, Tongyu Liu, Tong Sun and Kenneth T V Grattan

***Abstract*—In this paper, a stable and reliable CO monitoring system with high sensitivity (at sub-ppm level) was designed and demonstrated with particular reference to use in the mining industry, tailoring the design specifically for forecasting spontaneous combustion, a major hazard to miners. The design uses the familiar approach of direct absorption spectroscopy approach, but showing novelty in the tailoring of this approach to the mining industry, for use *in situ.* An appropriate strong CO absorption line is used to help eliminate the cross interferences expected from gases present in ambient air, with several preferred CO absorption lines selected and investigated, therefore choosing a distributed feedback (DFB) laser operating at a wavelength of 2330.18 nm as the excitation source. Through a detailed investigation, a minimum detection limit of ~0.2 ppm and a measurement precision (σ) of 6.3 ppb were achieved with a data updating rate of 6.6 s/point. Further in tests for the mining industry, a long-term continuous monitoring evaluation was carried out, demonstrated the excellent stability and reliability of the CO monitoring system. The results obtained have validated the potential of this design of a CO monitoring system for practical monitoring applications underground to enhance safety in the mining industry.**

***Index Terms*—carbon monoxide, mining industry, direct absorption spectroscopy, TDLAS**

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I. Introduction

As a colorless and odorless gas, carbon monoxide (CO) is rightly called the "silent killer" owing to its strong affinity for hemoglobin and even short-term exposure of humans to CO gas at high levels may result in irreversible brain damage [1]. Further, high levels of CO gas present in ambient air is very dangerous since it can burn and even explode, when the concentration ranges from 12.5% to 74.5%. Therefore, creating an effective CO monitor is important to be available for industrial process analysis and environmental pollution and human monitoring [2]-[5], especially for a range of practical safety-focused monitoring applications, such as forecasting spontaneous combustion in the mine goaf areas [6], [7]. Typical monitored concentrations of CO present to suit these applications range from 1 part per million (ppm) by volume in ambient air to 1000 ppm levels for industrial monitoring. Hence, there is a significant demand for an *in-situ* and cost-efficient CO monitoring system that demonstrates ultra-high sensitivity, long-term stability and reliability and is tailored for use in mines, to reduce hazards to miners.

Compared with other non-optical methods for gas detection, tunable diode laser absorption spectroscopy (TDLAS) technology is known to show real advantages and potential for real time monitoring, offering high selectivity and sensitivity, long-term stability and excellent reliability [8]-[10]. The principles of TDLAS-based CO sensors have been demonstrated by different groups, for example using a single mode distributed feedback (DFB) laser in the near-infrared region [11], [12], an interband cascade laser (ICL), or a quantum cascade laser (QCL) in the mid-infrared (MIR) spectral region [13]-[16]. Zhang *et al*. have developed a fiber-based CO sensor with a 1.56 μm DFB laser applied to the TDLAS method, achieving a detection limit of 5 ppm. Because of the advantage of intrinsic safety that it offers, such a sensor scheme has already been successfully applied in the field of coal mine safety [12]. In 2017, Ghorbani *et al*. presented a compact MIR CO sensor with a detection limit of 9±5 ppb by employing a room-temperature (RT) ICL emitting at 4.69 μm and a circular multi-pass gas cell (MGC) with a 4-m optical length [13]. In 2018, Dang *et al*. demonstrated a CO sensor by

the use of a RT continuous wave (CW) DFB QCL, operating at 4.76 μm and a 1.6-m mini MGC [16]. A minimum detection limit of 26 ppb was obtained with a 1 s acquisition time. Moving forward in this work from those developments and recognizing that ultra-sensitivity CO sensors have been reported, a CO sensor using a laser diode emitting at 2.3 μm can also provide highly sensitive detection, taking advantage of the stronger absorption of the CO first overtone band. Importantly, it can avoid the disadvantage that the very high cost that some MIR laser sources brings. Thus these 2.3 μm sources make possible the design of monitoring systems suited to a wide range of practical applications and especially for mine safety, the focus of this work, where the low cost can ensure wider use.

Such a sensor, employing a 2.3 μm laser diode, has attracted significant attention in recent years and a number of CO sensors have been reported and applied across a diverse group of industries [17]-[20]. Examples include a compact and calibration-free CO sensor with an in-line reference cell [21], a real-time *in-situ* CO sensor in a pulverized-coal-fired power plant [22], a ppm-level CO sensor for application to early fire detection [23] and a sensitive CO sensor used for SF_6 decomposition analysis in an electrical power system [24]. Among these methods, the technique of 2f wavelength modulation spectroscopy (WMS) was adopted in the reported CO sensors. However, as a spectroscopic method for gas detection, a further commonly used technique for TDLAS is termed direct absorption spectroscopy (or line scanning), which allows for a more reliable sensor, with a simple and compact system structure to be designed. Normally, direct absorption spectroscopy shows a worse detection sensitivity by two orders of magnitude than that of WMS. However, the detection sensitivity can be greatly improved by using fast-scanning direct absorption spectroscopy, accompanied by use of a signal averaging process.

In this paper, a stable, reliable and sensitive (sub-ppm range) CO monitoring system has been demonstrated, improving on previously reported designs and targeted on forecasting spontaneous combustion in the mining industry, by using the optimized direct absorption spectroscopy approach.

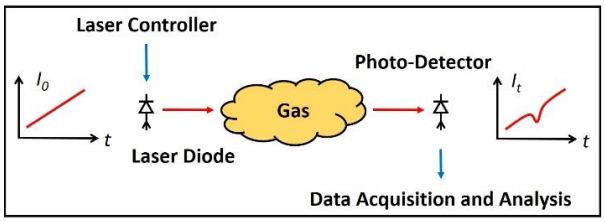

Fig. 1. Schematic diagram of the operation of a direct absorption optical sensor.

In optimizing this design, the CO absorption lines preferred to eliminate the cross interference from CO_2 and H_2O in ambient air were firstly investigated. A DFB laser operating at a specific wavelength of 2330.18 nm and a 20-m MGC were employed to target the strong absorption line of the CO first overtone band and to increase the effective optical length, respectively. After a careful investigation of the system performance, a minimum detection limit of ~0.2 ppm and a measurement precision (σ) of 6.3 ppb were achieved, with a data updating rate of 6.6 s/point. This design was evaluated using long-term continuous monitoring, to demonstrate the excellent reliability and stability of the CO monitoring system that has been developed.

II. Detection Methodology

A. Direct Absorption Spectroscopy

Figure 1 shows the schematic diagram of the approach chosen for a direct absorption based optical sensor for mining applications. By ramping up the laser diode injection current, the emitting laser wavelength can be tuned in a way that is approximately linear. The intensity decreases when the laser light is passed through the gas sample investigated and as a result, the small dip in the background line monitored creates the absorption signal due to the presence of the explosive gas. According to the Lambert-Beer law, the gas concentration can be deduced using the following formula [25]:

$$C = \frac{ln(I(\lambda)/I_0(\lambda))}{\alpha(\lambda)L} \quad (1)$$

$$\alpha(\lambda) = S(T)P\psi(\lambda) \quad (2)$$

where C is the volume concentration of the gas sample, λ is the laser wavelength, L is the length of the optical path, $I_0(\lambda)$ is the initial light intensity, $I(\lambda)$ is the transmitted light intensity due to the absorption, $\alpha(\lambda)$ is the absorption coefficient, P is the total pressure of the gas medium, $\Psi(\lambda)$ is the spectral line shape and $S(T)$ is the intensity of the characteristic spectral lines, which show a temperature dependence.

B. Preferred CO Absorption Line

Among the group of reported CO sensors, the use of laser diodes operating around 1.6 μm, 2.3 μm and MIR laser sources (QCL or ICL) near 4.6 μm have most frequently been employed to allow the emission to correspond to the CO absorption band of the weaker second overtone band, first overtone band and the strongest fundamental vibration band, respectively. The performance of these TDLAS-based sensors is strongly dependent on the strength of the gas absorption line. Thus, the selected laser sources must target the most suitable CO absorption line, ideally one with a high cross-section to enhance the sensitivity of sensor. Arising from their stronger absorption bands and yet being cost-effective to use, commercially available laser sources emitting around 2.33 μm are the preferred candidates to detect CO gas with a ppm-level sensitivity, ideal for the mining applications under consideration.

At the same time, it is equally important to eliminate the effect of cross interferences that may be seen from gases present in the ambient air. The potential for such cross interference effects mainly originates from the presence of different hydrocarbon gases, arising from their absorption bands overlapping with those of CO in the infrared spectral region. For a CO monitoring system, the spectral interference effects seen from both CO_2 and H_2O are the major challenges in ambient air, the consequence of which is to reduce the reliability of many monitoring systems in this application. In addition, methane (CH_4) is also an important interferening gas,

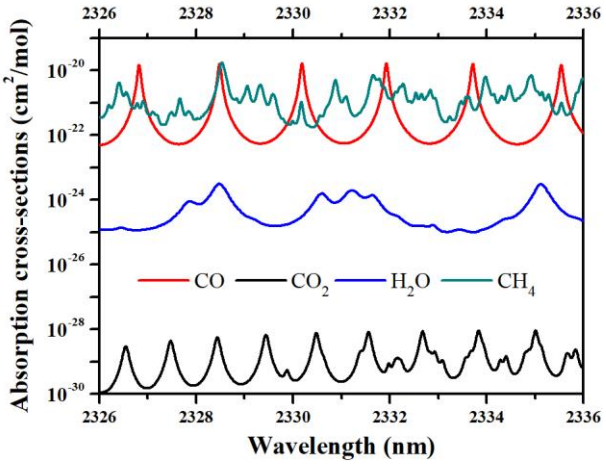

Fig. 2. The absorption cross-sections of CO, CO_2, H_2O and CH_4 at wavelength around 2.33 μm.

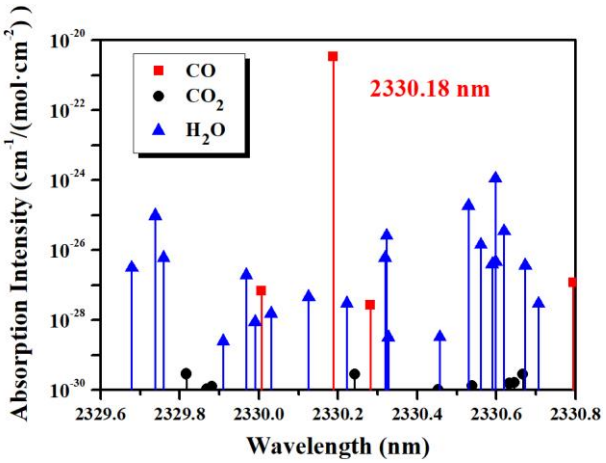

Fig. 3. Illustration of key absorption lines of CO, CO_2, and H_2O near 2.33 μm.

besides CO_2 and H_2O, for the application in mining industry. Figure 2 shows the absorption cross-sections of CO, CO_2, H_2O and CH_4 over a wavelength range from 2326 nm to 2336 nm, based on the Hitran 2012 database [26], where it can be seen that the absorption cross-section of CO is greater than that of CO_2 by nine orders of magnitude, showing that almost no interference from any pure CO_2 gas that may be present is seen. The wavelength of 2328.5 nm cannot be used for sensitive CO detection under a high humidity environment, because there are only four orders of magnitude of difference between the CO and H_2O absorptions, as shown in Figure 2 and inmnay practical mining cases there may be high levels of water vapor present. The absorption peak of CO at 2333.7 nm is a good choice to avoid such cross-interference effects from both CO_2 and H_2O. The absorption peak of CH_4 at 2330.2 nm is the same wavelength as that of CO, and the absorption cross section of CH_4 is obviously weaker than that of CO at 2330.2 nm, which makes this wavelength a good choice for the simultaneous detection of CO and CH_4 gas for applications in coal mines.

Given the above, a wavelength of 2330.18 nm was adopted as the basis of developing the optimum design of a CO monitor in this evaluation, recognizing both the line intensity available and the problem of cross interference effects. Figure 3 depicts the absorption lines of CO, CO_2, and H_2O near a wavelength of 2330 nm. As shown, the intensity of CO absorption line at a wavelength of 2330.18 nm is 3.39×10^{-21} $cm^{-1}/(mol\bullet cm^{-2})$ at room temperature and at atmospheric pressure. This is obviously a stronger absorption than that of the CO_2 and H_2O lines, which is good. Therefore it forms a sound basis for the development of a highly sensitive and selective CO monitor, using a laser diode operating at 2330.18 nm.

III. Experimental Investigation

A. Monitoring System Structure

Figure 4 shows a schematic of the CO gas monitoring system developed in this work for coal mine use, it consisting of three major components: an optical assembly, the laser controller and a signal acquisition and analysis system. For ease of construction, both commercially available and specially designed components to create the innovative, tailored design of the whole system developed are used. In the optical assembly, a single-mode fiber pigtailed CW DFB laser

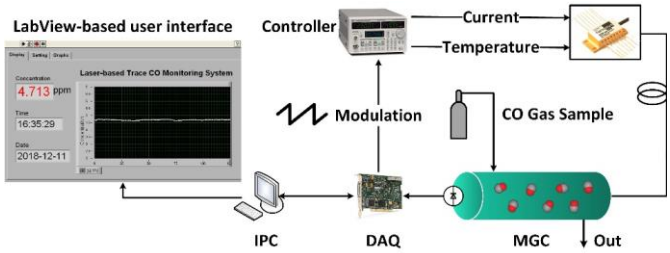

Fig. 4. Schematic of the CO gas monitoring system developed showing the major components. IPC: industrial personal computer, DAQ: data acquisition, MGC: multi-pass gas cell.

(Nanoplus GmbH) was employed as the excitation source, operating at a central wavelength of 2330 nm. The tuning coefficients of current and temperature were determined to be 0.02 nm/mA and 0.23 nm/ºC, respectively, enabling the fine control of the system that was required. The Herriott-type MGC has an effective optical length of 20 m, with dimensions of 314×89×104 mm^3 and a basic volume of 0.57 L. Both sides of the inside of the gas cell were dielectric coated, providing a broadband transmission over the wavelength range from 2290 nm to 2370 nm. The transmitted laser beams were detected by using the RT extended-InGaAs photo-detector installed within the system. Applying a stable current driver and temperature controller (Stanford Research Systems, LDC510) and using butterfly laser diode mounts, the DFB laser operated steadily at a temperature of 29.5ºC, generating an output power >2 mW. At the same time, the modulation of the laser wavelength was obtained using a sawtooth wave, over a current range from 60 mA to 90 mA. Following data acquisition (DAQ) and analog-to-digital conversion (National Instruments Corporation, PCI-6221), the detected signals were passed to the signal analysis system for further investigation.

B. LabVIEW-based Software

The absorption signals that were detected were processed using specially-developed LabVIEW-based programme. As depicted in Figure 4, the user interface for the CO monitoring system comprises three key parts: real-time data for the CO

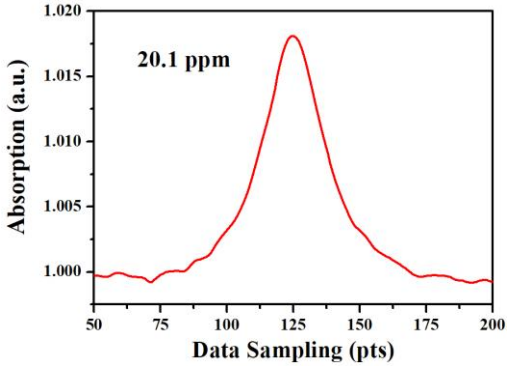

Fig. 5. Normalized signal obtained with a nominal CO gas at a concentration of 20.1 ppm.

concentration, the operating parameters of the system and the detected signals represented graphically. The system was designed to display to the user the key control parameters and the 'run data' from the CO detection, together with displaying the graphs showing the changes in the detected signals, the normalized signals, the nonlinear least-squares fitted curve and thus a real-time measured value of the detected CO concentration, such as would occur in the coal mine.

C. Data Processing Approach

In order to conduct real time *in-situ* detection of the concentration of the CO gas, the fast-scanned direct absorption spectroscopic approach, accompanied by use of an averaging process, was employed in the evaluation carried out. Initially the 2.3 μm laser diode was tuned using a 30 Hz sawtooth wave to scan repeatedly the laser wavelength around 2330.2 nm. A consecutive acquisition of 200 data points was carried out for each cycle of the spectrum. The top 50 data points of every spectrum acquired were discarded because of the strong fluctuations observed, this being due to the current jump seen in sawtooth wave. Following that, the spectra obtained were averaged successively and fitted by using a multi-order polynomial to allow for normalization of the averaged spectra. Finally, an algorithm giving nonlinear least-squares fitting was employed to remove random noise and reduce the potential etalon effect, and thus improve the signal-to-noise ratio (SNR) of system. In this work, a sample of CO gas at a nominal concentration of 20.1 ppm (and typical of actual coal mines) was employed, creating the consequent normalized spectral signature, which is depicted in Figure 5.

IV. Results and Discussions

A. Averaging Times of Spectra

The raw data updating rate of the CO monitor designed can be increased greatly with the use of a low averaging times for the spectra obtained. When the spectra were averaged successively 3 times, an updating rate for the data of ~0.1 s/point could be realized. However, the low averaging times of the spectra obtained will result in a larger level of data fluctuation and signal instability monitored, this being due to the white noise present. Therefore, the most suitable averaging times for the spectra were evaluated firstly, in that way to improve the stability of the CO monitoring. During the evaluation, the nominal 20.1 ppm CO gas was continuously filled into the MGC, at a flow rate of 1 L/min. The spectra obtained were averaged either 50, 100 and 200 times respectively. Figure 6 shows the corresponding CO concentration curves and the distribution histograms, each of which contain 100 raw data points. As depicted in Figure 6, the consequent frequent fluctuations in the CO concentration can be observed with the lowest averaging times used (50), which resulted in an apparently wider range of concentration distribution being determined. However, the frequency of the fluctuations observed decreased when using the 100 times averaging. Finally, when the 200 times averaging was used, the stability of the monitored CO signal improved significantly. A Gaussian profile was employed to fit the distribution histograms and as a result the *R*-square value, the half-width at half-maximum (HWHM) and the sigma value (σ) were calculated to be 0.9949, 14.8 ppb and 6.3 ppb, respectively. The result demonstrated the excellent stability and precision obtainable from the use of the CO monitoring system developed. Given that result, the 200 times averaging was employed in the rest of this work, corresponding to a data updating rate of 6.6 s/point.

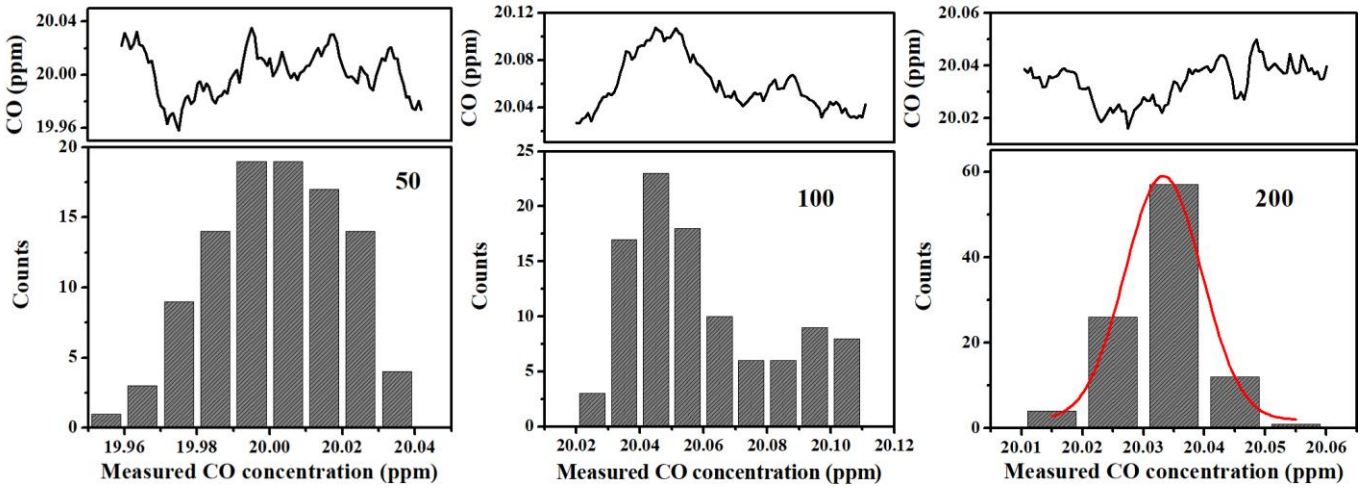

Fig. 6. The CO concentration curves (top) and the distribution histograms (bottom) with different spectral averaging times (left to right: 50, 100 and 200).

B. *Performance of the CO Monitoring System*

To evaluate further the performance of the CO monitoring system developed, the detection sensitivity and the linear

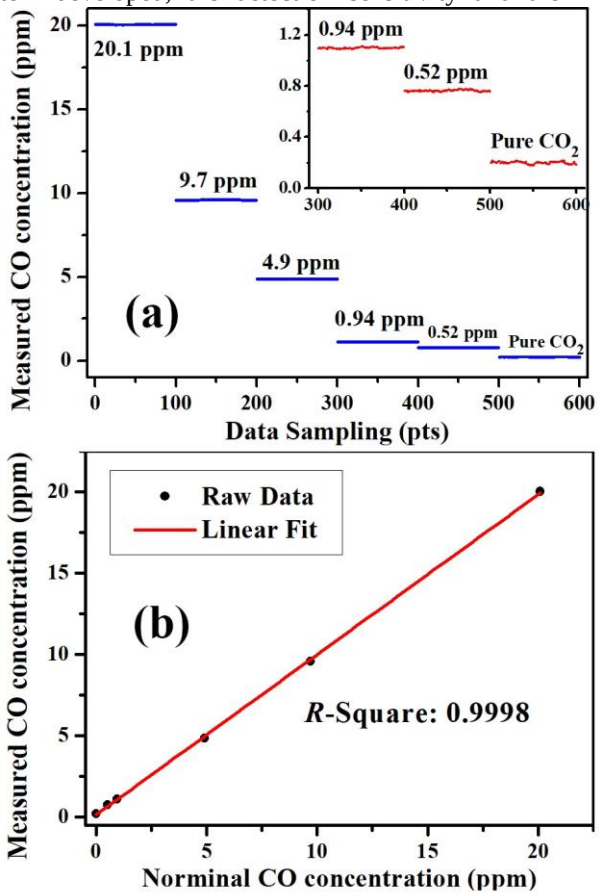

Fig. 7. (a): measured CO concentrations determined from the monitoring system developed, (b): graph of the measured CO concentration versus the nominal CO concentration used.

concentration response were evaluated in a series of experiments carried out. Various different CO gas samples at different nominal concentrations of 20.1 ppm, 9.7 ppm, 4.9 ppm, 0.94 ppm and 0.52 ppm were used, the samples being successively filled into the MGC (with a constant flow rate of 1 L/min). 100 sample points to monitor the CO concentration were recorded, for each concentration level and results are depicted in Figure 7(a). The CO monitoring system exhibited excellent detection sensitivity, precision and stability. As shown from the inset in Figure 7(a), the CO monitoring system could readily sense the presence of CO gas, at a concentration as low as 0.52 ppm, with excellent stability. Next the system was evaluated with pure CO_2 gas (99.999%), which was filled into the MGC to verify the real 'zero baseline' of the monitor. Given that such a CO_2 sample should produce no interference in the spectra recorded, the data monitored and depicted in Figure 7(a) can then be used to determine that the detection limit of system was below 0.2 ppm. The measured data were averaged for each concentration level and then plotted as a function of the nominal CO concentrations used, seen in Figure 7(b). The *R*-square value was calculated to be 0.9998 with a linear fitting analysis, which demonstrates the excellent linear concentration response of the monitoring system developed in this work.

C. *Long-Term Continuous Monitoring*

Long-term stable and reliable operation of a CO sensor is of great significance for monitoring across a range of practical

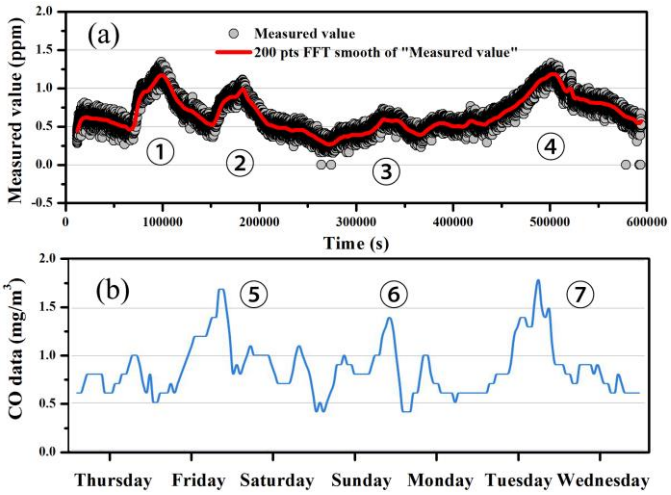

Fig. 8. The measured CO concentration determined (a) using the monitoring system developed, (b) compared with the official monitoring data from the detection system installed by the Jinan Government.

applications, especially for this application in mining industry. Thus in order to test the stability of the CO monitoring system developed, the system was installed and operated continuously for just over a week. Figure 8(a) shows the data recorded over ~170 hours of use, in an ambient atmosphere. The raw data were processed using the 200 pts FFT smoothing method, in order to reveal the key trends more clearly. The results show that there are four peaks, illustrated in Figure 8(a), which are labelled in sequence. Looking at these closely, the first peak (1) resulted from the leaking CO gas from another experiment being carried out in the same place. Three other peaks (labelled (2), (3), and (4)) reflect the actual fluctuations of the CO gas concentration in the ambient air. To demonstrate the high performance of the CO monitoring system developed, Figure 8 contrasts the results obtained from the monitor developed in this work with the official monitoring data for CO gas concentration, (from an official monitor installed by the Jinan government). As shown in the Figure 8, the three peaks, labelled (2), (3), and (4) are consistent with the peaks labelled (5), (6), and (7) which were obtained in the official monitoring exercise carried out over one week of operation and there is good agreement seen (except for the 'rogue peak' (1)). This result has clearly demonstrated the long-term stability and reliability of the monitoring system developed in this work for coal mining applications, showing the positive feature that there is no cross interference seen in the monitoring of the ambient air, simplicity of operation and affordable cost.

V. Conclusions

In summary, a reliable CO monitoring system with high sensitivity (sub-ppm level) and good long-term stability was

designed, developed and evaluated, using a 2.33 μm laser diode and employing the optimized direct absorption spectroscopy technique. Compared with the use of lasers at wavelengths of 1.6 μm and 4.6 μm, the commercially-available laser sources used, operating at 2.33 μm, are the preferred candidates for the effective detection of the CO gas, showing the advantage of being cost-effective to monitor the stronger absorption of the CO first overtone band. The use of fast-scanning direct absorption spectroscopy, accompanied by an optimized averaging process, has allowed the development of a sensor system that is relatively inexpensive, yet reliable and sensitive, having a simple and compact system structure and well suited to the needs of today's mining operations. In order to eliminate cross interference from other gases present in the ambient air and target a strong CO absorption feature, the selection of the most suitable CO absorption line was investigated in this work. As a result, a DFB laser operating at a specific wavelength of 2330.18 nm was determined to be the optimal excitation source to use. After a careful investigation of the system thus designed and constructed, its performance using the spectral data processing approach was evaluated and a minimum detection limit of ~0.2 ppm and a measurement precision (σ) of 6.3 ppb were achieved, (with a data updating rate of 6.6 s/point). Long-term continuous monitoring using the system has demonstrated its excellent stability and reliability for use in China's coal mines. A good comparison with 'official data' from a Government-approved monitoring system was obtained *in situ*, thereby validating such a CO monitoring system as a practical means of monitoring in mines and thus for forecasting situations where spontaneous combustion, hazardous to miners, could occur.

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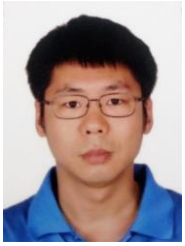

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