OPTOELECTRONICS LETTERS Vol.11 No.4, 1 July 2015

A mid-infrared methane detection device based on dual-channel lock-in amplifier*

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(Received 28 May 2015)

A portable dual-channel digital/analogue hybrid lock-in amplifier (LIA) is developed, and its amplitude detection error is less than 10% when the signal-to-noise ratio (SNR) is larger than −12 dB. Then, a differential mid-infrared methane (CH_4) detection device is experimentally demonstrated based on a wideband incandescence wire-source and a multi-pass spherical reflector. The experiments are carried out to obtain the sensing performance of the device. With the absorption length of only ~4.8 cm, the limit of detection (LoD) is about 71.43 mg/m^3, and the detection range is from 0 mg/m^3 to 5.00×10^4 mg/m^3. As the concentration gets larger than 714.30 mg/m^3, the relative detection error falls into the range of −5%—+5%. Two seven-hour-measurements are done on the CH_4 samples with concentrations of 1.43×10^3 mg/m^3 and 4.29×10^3 mg/m^3, respectively, and the results show that the maximum relative error is less than 5%. Because of the cost effective incandescence wire-source, the small-size and inexpensive dual-channel LIA, and the small-size absorption pool and reflector, the developed device shows potential applications of CH_4 detection in coal mine production and environmental protection.

Document code: A **Article ID:** 1673-1905(2015)04-0298-5

DOI 10.1007/s11801-015-5102-2

As a primary technology, infrared (IR) spectroscopy absorption has been widely used in methane (CH_4) measurement[1-4]. Existing IR CH_4 sensors can be classified into three types, namely, photoacoustic spectroscopy (PAS)[4], wavelength modulation spectroscopy (WMS) or tunable diode laser absorption spectroscopy (TDLAS) based on distributed feedback lasers (DFBLs)[5-7] and direct absorption spectroscopy (DAS)[8]. Recently, much effort has been devoted to CH_4 detection by our group (Jilin University, China), and several detection systems or devices based on TDLAS and DAS techniques have been developed[9-12]. During the design and development of CH_4 detection sensors, noises and interferences from electrical components and wires as well as environmental changes are important factors which can deteriorate the detection performance. As is well known, a lock-in amplifier (LIA), either digital-type or analogue-type, is an efficient and important tool to extract sensing information from highly polluted signal, which is widely used in sensor applications[13-17]. However, since the commercially available LIA is too expensive in price and too large in size, it cannot be integrated into a portable CH_4 detection device. Therefore, as an obvious difference from our previous reports[9-12], in this paper, we develop a dual-channel digital/analogue hybrid LIA for suppressing the noises in the two IR sensing signals, which can be integrated in the mid-IR CH_4 detection device.

The structure of the detection sensor is shown in Fig.1, which generally contains four modules, namely the control and processing module, the gas cell and reflector, the signal pre-processing module and the dual-channel LIA. An ARM7 processor (LPC2148, Philips, Holland) is used as the primary control and processing component. A spherical mirror is used as absorption pool as well as light reflector with absorption length of 4.8 cm. A wide-band IR light-source (IRL715, PerkinElmer Optoelectronics, Germany) and dual-channel IR detector (LMM-242, Infra Tec, Germany) equipped with two optical filters centered at 3.31 μm and 3.95 μm are located at the upper and lower sides of the center, respectively. The 4 Hz square wave modulation signal processed by a voltage control constant current source (VC-CCS) circuit at ON state is supplied to the IR light-source.

The sensor contains two channels. For the detection channel, the light reaching the λ_1=3.31 μm filter can be absorbed by CH_4 inside the gas-cell, and the detection electric signal $u(\lambda_1,t)$ is produced through IR detector. For the reference channel, the light reaching the

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* This work has been supported by the National Natural Science Foundation of China (No.61307124), the Changchun Municipal Science and Technology Bureau (No.14KG022), and the Education Department of Jilin Province in China.

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λ_2=3.95 μm filter cannot be absorbed by CH_4, and the reference electric signal $u(\lambda_2,t)$ is produced through the IR detector. These two electric analogue signals are both processed by pre-amplifier (PA) and signal adjuster (SA). Then the two signals propagate into a substractor (ST), and a differential signal (DS) is generated, i.e., $\delta u(t)=u(\lambda_2,t)-u(\lambda_1,t)$. The differential signal $\delta u(t)$ and the reference-channel signal $u(\lambda_2,t)$ are further processed by a low-pass filter (LPF), an LIA and a sampling/holding (S/H) module, and they are finally converted to digital signals via a dual-channel 16-bit analog-to-digital converter (ADC). Each branch of the dual-channel LIA is composed of a multiplier (MUL), a shift-phase module (SPM), an LPF and an SA.

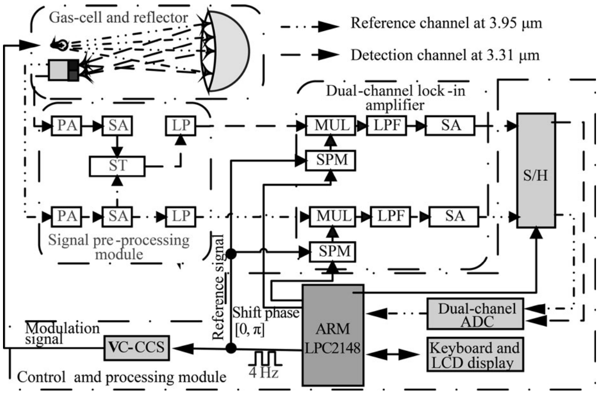


Fig.1 Configuration of the mid-IR CH_4 sensor, containing the control and processing module, the gas cell and reflector, the signal pre-processing module and the dual-channel lock-in amplifier

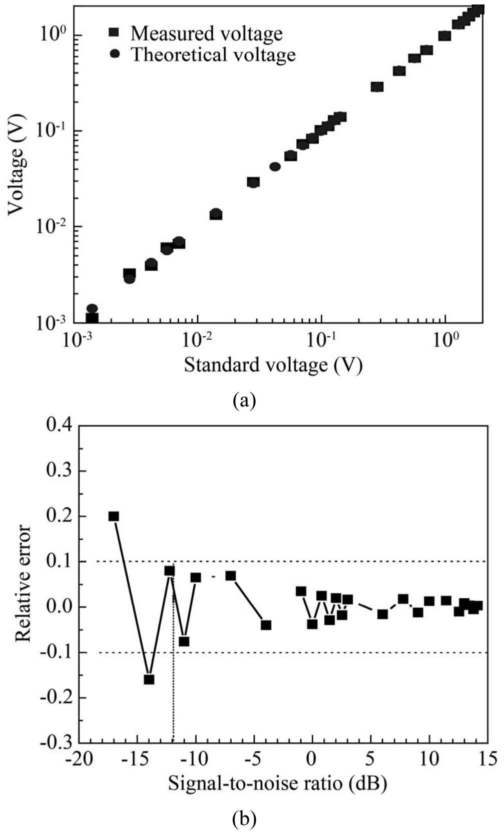
Though there are various commercially available LIAs, they cannot be integrated in the detection device due to the high cost and the large size. Therefore, a portable dual-channel LIA is developed with low cost and normal performance, whose structure is shown in Fig.1. The LIA involves SPM, MUL, LPF and SA. The 4 Hz reference square wave signal generated by ARM processor is supplied to SPM, and its phase can be tuned from 0° to 180°. A high precision balanced modulator (AD630), which combines a flexible commutating architecture with accuracy and temperature, is used to realize the multiplication between the differential signal $\delta u(\lambda_1, \lambda_2, l, t)$ (or reference-channel signal $u(\lambda_2,t)$) and the reference phase-shifted 4 Hz square-wave signal $r(t)$. The output signal from AD630 is processed by an 8th-order, low-pass and switched-capacitor Butterworth filter (MAX291). Finally, the signals are adjusted for the process by the sampling and holding (S/H) module.

Let the standard cosine wave signal, whose amplitude changes from 0.12 V to 0.54 V, input into one branch of the LIA. Tuning the phase difference between the cosine wave signal and the reference signal to be 0°, we measure the direct current (DC) voltage output from the LIA. The obtained fitting equation between the theoretical voltage (defined as x) and the measured voltage (defined as y) is

$$y = 1.041\,48x - 0.002\,47\,. \tag{1}$$

The linearity degree of fitting is 99.978%, which indicates that the LIA has high linearity.

Normally, the detection limit of the LIA is dominated by the electronic noise of the hardware system. When the amplitude of the pure cosine wave signal is increased from 0.02 V to 2.6 V, the measured DC voltage output from the dual-channel LIA and the theoretical standard voltage are both shown in Fig.2(a) as a clear comparison. We then calculate the signal-to-noise ratio (SNR) and the relative error of the DC output voltage compared with the theoretical value, and the relation is shown in Fig.2(b). As can be seen, once the SNR of LIA is less than −12 dB, the relative error of LIA is larger than 10%, which means that the amplifier cannot be used to make precise measurement on signals with SNR less than −12 dB. So the minimum SNR of the LIA is determined to be −12 dB.



(a)

(b)

Fig.2 (a) The measured DC voltage output from one branch of the LIA and the required theoretical standard voltage; (b) Curve of the relative error between the standard voltage and the measured voltage versus SNR

A glass container with volume of 50 L is used for preparing CH_4 samples and serves as testing environment. For the container full of pure N_2, proper amount of pure CH_4 is injected into it via an injection needle, and standard gas samples with concentration range from 0 mg/m^3 to 5.00×10^4 mg/m^3 are prepared. Under different concentrations, the two amplitudes $\delta U(\lambda_1, \lambda_2, l)$ and $U(\lambda_2, l)$ are sampled, and the ratio $\delta U(\lambda_1, \lambda_2, l)/U(\lambda_2, l)$ is calculated. The experimental data are shown in Fig.3. We then obtain their fitting relation as

$$\frac{\delta U(\lambda_1,\lambda_2,l)}{U(\lambda_2,l)} \approx 0.269\,97\exp\left(-\frac{C}{30\,199.5}\right)+0.996\,48 , \tag{2}$$

which agrees with Beer-Lambert's law, and can also prove the correction of this experiment. Let $C=f(\delta U/U)$, and then we can get the formula of C as

$$C=-30\,199.5\times \ln\left[\frac{1}{0.269\,97}\left(\frac{\delta U(\lambda_1,\lambda_2,l)}{U(\lambda_2,l)}-0.996\,48\right)\right], \tag{3}$$

which can be used to determine the CH$_4$ concentration with the obtained ratio of $\delta U(\lambda_1, \lambda_2, l)/U(\lambda_2, l)$.

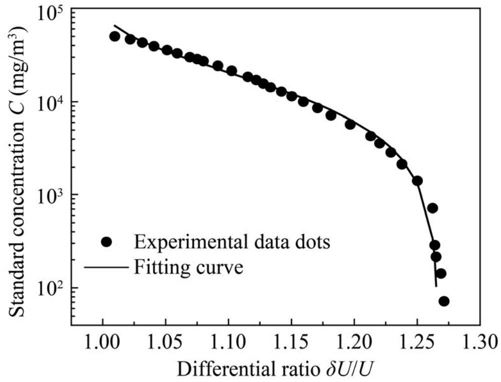
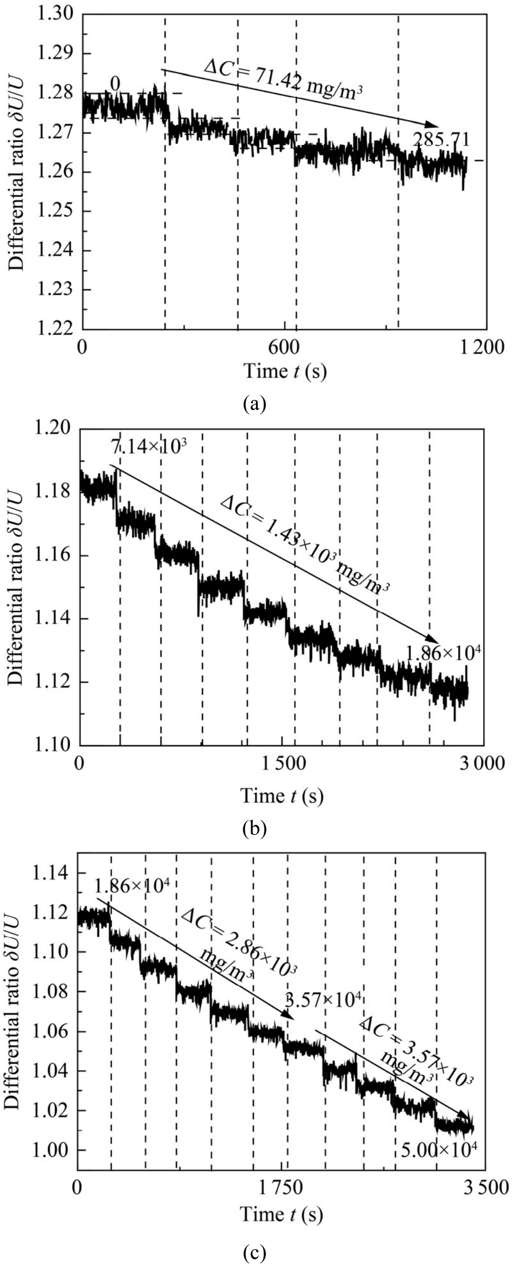


Fig.3 Experimental data and fitting curve of gas concentration C versus the differential ratio of $\delta U(\lambda_1, \lambda_2, l)/U(\lambda_2, l)$ in the concentration range from 0 mg/m^3 to 5.00×10^4 mg/m^3

The detected ratio ($\delta U(\lambda_1, \lambda_2, l)/U(\lambda_2, l)$) under certain concentration is not a constant, and the limit of detection (LoD) and sensitivity mainly depend on the noise level of the system. At an initial concentration, the detection sensitivity can be regarded as the minimum CH$_4$ concentration change which can be obviously recognized by the system. The gas sample with an initial concentration is firstly prepared, and then the concentration is slightly modified by injecting proper amount of pure CH$_4$ until the minimum detection amplitude is steadily lower than the maximum detection amplitude under the initial concentration. Fig.4 shows the measured ratios of $\delta U(\lambda_1, \lambda_2, l)/U(\lambda_2, l)$ in low concentration range of 0–285.71 mg/m^3, middle concentration range of 7.14×10^3–1.86×10^4 mg/m^3 and high concentration range of 1.86×10^4–5.00×10^4 mg/m^3. It can be found from Fig.4(a) that the LoD is about 71.43 mg/m^3, and the sensitivity within the low concentration range is also about 71.43 mg/m^3. It can be found from Fig.4(b) that the sensitivity in the middle concentration range of 7.14×10^3–1.86×10^4 mg/m^3 is less than 1.43×10^3 mg/m^3, since the ratio $\delta U(\lambda_1, \lambda_2, l)/U(\lambda_2, l)$ can be obviously distinguished from each other when the concentration is increased or decreased by 1.43×10^3 mg/m^3. From Fig.4(c), the sensitivity in the high concentration range of 1.86×10^4–3.57×10^4 mg/m^3 is less than 2.86×10^3 mg/m^3, and when the concentration is larger than 3.57×10^4 mg/m^3, the sensitivity is lower than 3.57×10^3 mg/m^3. It is also confirmed that the value of the sensitivity increases as the concentration gets larger.



(a)

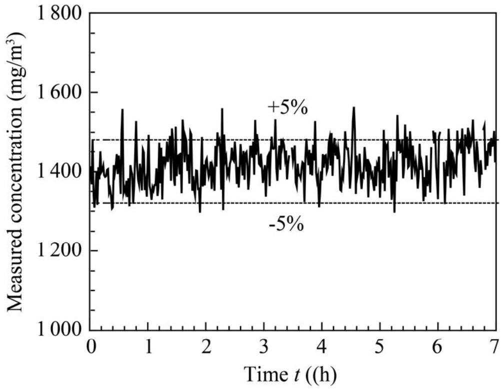
(b)

(c)

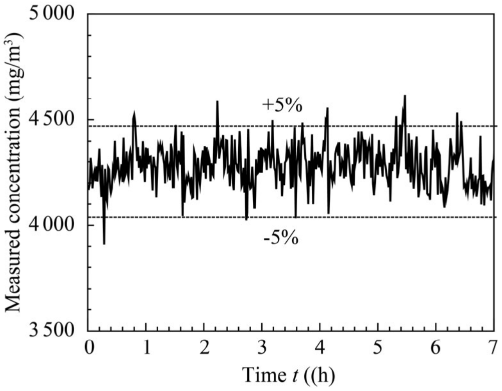
Fig.4 The measured differential ratios of $\delta U(\lambda_1, \lambda_2, l)/U(\lambda_2, l)$ in (a) low concentration range of 0–285.71 mg/m^3 with concentration sensitivity (ΔC) of 71.43 mg/m^3, (b) middle concentration range of 7.14×10^3–1.86×10^4 mg/m^3 with ΔC of 1.43×10^3 mg/m^3, and (c) high concentration ranges of 1.86×10^4–3.57×10^4 mg/m^3 with ΔC of 2.86×10^3 mg/m^3 and 3.57×10^4–5.00×10^4 mg/m^3 with ΔC of 3.57×10^3 mg/m^3

Because of noises and interferences introduced by the differential signal and the ratio signal, the detection stability may be affected. For the two prepared gas samples with concentrations of 1.43×10^3 mg/m^3 and 4.29×10^3 mg/m^3, the detection lasting 7 h is carried out on each sample, and the results are averaged per one minute. For the gas sample with concentration of 1.43×10^3 mg/m^3, the measured concentration range is roughly from 1.36×10^3 mg/m^3 to 1.50×10^3 mg/m^3, indicating a relative error of less than 5%, which is shown in Fig.5(a). For the gas sample with concentration of 4.29×10^3 mg/m^3, the measured concentration range is basically from 4.07×10^3 mg/m^3 to 4.50×10^3 mg/m^3, indicating an absolute error of less than 5%, as shown in Fig.5(b).

The performance of the mid-IR detection device at 3.31 μm using LIA is compared with those of our previously reported detection devices using least-square fast-transverse-filtering (LS-FTF) self-adaptive denoising algorithm[9] and wavelet-denoising (WD) algorithm[12], respectively. The comparison results are shown in Tab.1. For two previously reported devices, due to the adopted efficient denoising algorithms for extracting useful sensing information, their LoDs are as low as 5.71 mg/m^3 and 0.71 mg/m^3, respectively. In this paper, the reflective distance between light source and detector is as small as 4.8 cm, and the LoD is about 71.43 mg/m^3 for a wide detection range (DR) of 0–5.00×10^4 mg/m^3. Despite this, smaller detection error (±5%) is also achieved for this device compared with those of the two sensors reported in Refs.[9] and [12].



(a)



(b)

Fig.5 Results of seven-hour-measurements on the standard CH_4 samples with concentrations of (a) 1.43×10^3 mg/m^3 and (b) 4.29×10^3 mg/m^3

Tab.1 Comparison among our three developed mid-IR and near-IR CH_4 detection devices

Method	Wavelength (μm)	Line strength (cm per molecule)	Source	Gas-cell (Distance between light source and detector)	LoD (mg/m^3)	Error (%)	DR (×10^3 mg/m^3)
Ref.[9]	3.31 (Mid-IR)	10^{-19}	Wire-source	Symmetric ellipsoid multi-path gas-cell (7.5 cm)	5.71	±5%	0–28.57
Ref.[12]	1.654 (Near-IR)	10^{-21}	DFBL	Direct-incident gas-cell (20 cm)	0.71	±3.8%	0–35.71
This paper	3.31 (Mid-IR)	10^{-19}	Wire-source	Reflection mirror (4.8 cm)	71.43	±5.0%	0–50

In summary, a portable dual-channel digital/analogue hybrid LIA is experimentally proposed for effectively extracting sensing signal from the severely polluted signal, which can make precise measurement on the small signal with an SNR of less than −12 dB. Based on the dual-channel LIA, a differential mid-IR CH_4 detection device is developed by employing a wideband wire-source and a spherical multi-pass reflector. With the absorption length of only 4.8 cm, the LoD is about 71.43 mg/m^3, and the detection range is from 0 mg/m^3 to 5.00×10^4 mg/m^3. The maximum relative errors of seven-hour-measurements on the CH_4 gas samples with concentrations of 1.43×10^3 mg/m^3 and 4.29×10^3 mg/m^3 are both less than 5%. Due to the cost effective incandescence wire-source, the self-developed small-size and inexpensive dual-channel LIA, and the small-size absorption pool and reflector, the developed standalone device shows potential applications of CH_4 detection in coal-mine environment.

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