A Compact, Cylindrical Multi-pass Cell for Sensitive Detection of Gas Absorption

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We developed a compact trace gas sensor, which incorporates a cylindrical thin-disk multi-pass cell. This cell is useful because the optical path length can be selected easily in a small absorption volume across the beam path of 25 cm³. We studied the optical characteristics of this cell and demonstrated some gas measurements with this system. This paper discusses the performance of the gas detection system which incorporates a cylindrical cell.

Key Words: Laser spectroscopy, Cylindrical multi-pass cell, Gas monitor

1. Introduction

The demand for compact trace gas detectors is increasing in many application areas including environmental science, medical diagnostics, and industrial process control and so on. Sophisticated absorption-based spectroscopic techniques such as modulation spectroscopy, cavity ring down spectroscopy, and photo-acoustic spectroscopy have been actively studied. For most laser absorption spectroscopic devices, in which a multi-pass cell is one of the key components, a variable and long optical path length (more than 10 m) and a small cell volume are desirable. There are two kinds of conventional cells, the White cells¹⁾ and Herriot cells.²⁾ Optical length of White cells can be varied; however, they have a large cell volume. On the other hand, Herriot cells have a small cell volume; however their optical path length cannot be changed easily. In this paper, we report the performance characteristics of a newly proposed cylindrical thin-disk-shaped multi-pass cell that has a variable optical length with a small cell volume.

2. Experiments

2.1 Cylindrical multi-pass cell

Figure.1 shows the cylindrical multi-pass cell. Spherical mirrors that are divided into six segments are placed in a circle. The mirror radius is 80 mm. The incident light is reflected in a star configuration, and comes out when the number of reflections equals a certain number (Fig. 2). The absorption volume across the beam path is 25 cm³. As a result of this optical design, the optical path length of the cylindrical cell can be changed easily by changing the incidence angle. The relation between the incident angle and the number of reflections is given by

$$(180 + 2\theta) \times N = 360 \times m \quad (1),$$

$$(N = 2(m-1): \text{even} \quad N = 2m-1: \text{odd})$$

where θ is the incidence angle and N is the number of reflections. The path length changes as a function of θ as

shown in Fig. 3.

When the incidence angle becomes smaller, the optical path length becomes longer. However, the beam expands with an

Fig. 1 Cylindrical multi-pass cell. Mirror radius is 80 mm. The optical path length can be easily changed between 0.4m and 10m.

increase in the number of reflections because of aberration. So,

Fig. 2 Optical path trace of cylindrical cell. Incident light is reflected in a star configuration. The beam diameter could be conserved for long path lengths more than 10 m.

Fig. 3 Relation between the input angle and optical

Fig. 4 Profile images of the input and output beams.

optical path length is limited. Therefore, we optimized the mirror positions on the basis of the analysis of the beam propagation, and the beam diameter was conserved for long optical path lengths of more than 10 m. Input and output beam profile images are shown in Fig. 4.

2.2 Experimental set-up

A schematic of the experimental set-up is shown in Fig. 5. The set-up consists of a light source with trace gas detection and gas handling systems. We used three kinds of lasers in three respective measurements. To measure CO_2 and NH_3 gas mixture absorption spectra in the near-IR, a DFB-LD (NTT Electronics Inc., NLK1554STB) was operated at a wavelength of 1537 nm with an output power of 15 mW and a linewidth of less than 1 MHz. To measure CO_2 absorption in a wide range, a widely tunable laser (Koshin Kogaku Co., Ltd LS-601A-56S2) was operated from 1525 nm to 1630 nm with an output power of 1 mW and a linewidth of less than 1 kHz. The wavelength resolution was less than 0.1 pm. To measure NO_2 absorption, we used periodically poled lithium niobate (PPLN) green (532 nm) laser. Each light source was aligned and passed through the cylindrical cell. The output beams were focused on photovoltaic semiconductor detectors by a

Fig. 5 Schematic of the experimental set-up.

gold-coated off-axis parabolic mirror with a focal length of 50.4 mm. The detected signal was acquired by a laptop computer with a 16-bit A/D data acquisition card (National Instruments, DAQ card-AL-16XE-50), and the signal data were analyzed using LabVIEW software. When we used DFB-LD, the absorption data were typically accumulated for 3-5 s, corresponding to 600-1000 frequency scans. A specific gas mixing system was designed to simulate flue gas conditions. The gas handling system consisted of gas input, an optical cell, and vacuum stages. Each gas bottle containing a sample was connected to a gas bottle containing pure N_2 for dilution. We could control the concentration and pressure of each sample. All gas handling lines were fabricated from stainless steel tubing.

3. Results and Discussion

3.1 Measurement of CO_2 and NH_3 absorption spectra at 1537 nm

Figure 6 shows CO_2 and NH_3 gas mixture absorption spectra around 1537 nm. The concentrations of the CO_2 and NH_3 gas samples were adjusted individually by mixing with pure N_2 gas. We used three concentrations for each gas. The partial pressures of CO_2 and NH_3 were both 50 Torr. The measurement temperature was 300 K, and the optical path length was 5.6 m.

Although the absorption cross section of CO_2 is very small in this range, we could detect this absorption. The absorption cross sections of CO_2 and NH_3 are 2×10^{-24} and $6 \times 10^{-22} \text{ cm}^2/\text{molec}$, respectively. We estimated the concentrations from these result (see Table 1). Ripple noise in the multi-pass cell was not completely removed, and the intensity of this ripple noise determines the measurement limit. The detection limit of CO_2 was 0.1%, and that of NH_3 was 30ppm.

Fig. 6 CO_2 and NH_3 gas mixture absorption spectra around 1537 nm. We measured three concentrations in both cases. Partial gas pressure was 50 Torr each. Optical path length was 5.6 m.

Table 1 Estimated concentrations of CO_2 and NH_3 .

	blue	red	black
CO_2	53.8%	74.4%	95.2%
NH_3	200 ppm	300 ppm	470 ppm

Fig. 7 CO₂ absorption spectra over a wide range. Experimentally observed spectra (black) and calculated ones (red) by HITRAN database. CO₂ gas pressure was 200 Torr. Optical path length was 11 m.

3.2 Measurement of CO₂ absorption over a wide range

Figure 7 shows CO₂ absorption spectra over a wide range. We measured from 1525 nm (6557 cm⁻¹) to 1630 nm (6135 cm⁻¹) continuously. The concentration of CO₂ was 100% and gas pressure was 200 Torr. The measurement temperature was 300 K. The optical path length was 11 m. The observed spectra are shown in black, and the calculated ones by HITRAN database are shown in red. The observed and calculated spectra are in good agreement.

We can see the ripple noise. The magnitude of the ripple noise depends on the optical path length. It may be generated by interference between the light scattered on the mirror and the output light.

3.3 Gas trace measurement of NO₂ at 532 nm

We used this system as a gas monitor. Figure 8 shows NO₂ gas trace spectra at 532 nm. We measured NO₂ at atmospheric pressure and at room temperature. The optical path length was 4 m. Gases passed in to the cell were N₂ or NO₂, and was changed after a certain time.

In this system equipped with a cylindrical cell, the measurement area has a very small volume of 25 cm³, making it sensitive to the change in the concentration of gas. The response time was 30 s. The measurement limit was 1.9 ppm estimated by the signal-to-noise ratio

Fig. 8 NO₂ gas trace spectra at 532 nm. NO₂ gas was injected and afterwards N₂ gas was injected at atmosphere pressure. Optical path length was 4 m.

4. Conclusion

We investigated the performance of a cylindrical multi-pass cell. The volume of the cylindrical cell is as small as a conventional Herriot cell and its optical length could be changed easily from 0.4 m to more than 10 m. The cylindrical cell has advantages over conventional cells. By measuring the absorption spectra, the performance of the cylindrical multi-pass cell were found to be comparable to that of a conventional Herriot cell. These results show that the performance of the cylindrical multi-pass cell is equivalent or superior to that of a conventional absorption cell. Specifically, it is compact and its optical path length can be easily varied.

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