Mid-Infrared light Source for ¹⁴CO₂ detection

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1. Introduction

In many nuclear facilities, radioactive compounds are released into the environment in gaseous form. In particular, C-14 is dominantly produced by a (n,α) reaction on oxygen-17 contained in water. The amount of C-14 is produced, in the case of the Wolsong nuclear power plants, at approximately 20 TBq/yr and around 1% of C-14 is released to the air in the form of carbon dioxide [1]. The emission requirement of carbon dioxide in Korea is less than 1 x 10⁴ Bq/m³. The carbon is the base material for the living organics, and it has been a worldwide trend to strengthen the monitoring of C-14 radioisotope.

A radiation detection technique is a general method to detect ${}^{14}CO_2$ in gaseous effluents. It is used to capture the carbon dioxide and detects the radiation from C-14 by a liquid scintillation counter [2]. It requires capturing and detection times and chemical processing steps to remove other radiation sources. This feature makes it difficult to detect ${}^{14}CO_2$ gas in real time.

The laser based detection technique, which is used to detect a unique absorption molecular spectrum, is one of the methods applied to real-time detection. However, it is hard to detect ¹⁴CO₂ using laser-based detection techniques, because the absorption cross sections are too low in the visible and near-IR regions, which are conventionally well developed. In recent researches, the corresponding wavelength region is moving to the mid-infrared region where the absorption cross sections become $10^5 \sim 10^6$ times bigger than those of the near-IR region. Thus, the detection of ¹⁴CO₂ in gaseous CO₂ becomes achievable even if there are some technical problems to obtain optics and detectors.

In this paper, we introduce the detection mechanisms for ${}^{14}CO_2$ and mid infrared light source generation.

2. Absorption Spectroscopy in Mid IR Region

Carbon dioxide has various vibrational levels originated from the stretching and bending between two oxygen atoms and one carbon atom, and rotational levels. The spectra consist of fundamental, overtone, and combination bands which are usually found in the infrared region. Spectroscopic tools, such as lasers, optics, and detectors, are well developed in the near infrared region and which is why the detection of molecules has been actively studied in the near-infrared and visible regions. However, in many cases, it is hard to detect low abundant molecules such as ${}^{14}CO_2$, because the absorption strengths are very low in the near-infrared and visible regions compared to the mid-infrared region. Since the absorption cross sections in the mid-IR region are an order of $10^5 \sim 10^6$ larger than that of the near-infrared region, the spectroscopic studies have moved to the mid-IR region for detecting trace molecules [3].

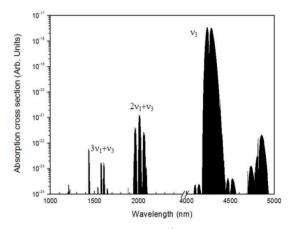


Fig.1. Absorption cross section for ¹²CO₂ molecule [3].

Figure 1 shows the ${}^{12}CO_2$ molecular spectra from the HITRAN database in the near-infrared to the mid-infrared regions.

3. Mid-IR source Generation

The cavity enhancement technique which improves the detection sensitivity, needs both easy frequency control and good beam quality, and thus we consider the difference frequency generation (DFG) as a frequency conversion for a mid-IR source at $4.2 - 4.5 \mu m$ [4].

2.1 Mid IR generation by using difference frequency generation

In this experiment, we used a MgO-doped periodically poled congruent lithium niobate (PPCLN) crystal for the frequency conversion which is available in the 4 μ m region. For mid-infrared generation with DFG, the 855 nm and 1056 nm beams co-propagated with vertical polarizations and focused on the MgO:PPCLN crystal in a single pass configuration. The crystal was mounted on an oven for the temperature phase matching. In the results, we obtained mid-IR light

at a wavelength of around 4.2 μ m to 4.5 μ m. The power was measured to be about 200 μ W depending on the wavelength and the incident laser power.

2.2 Linear absorption in the CO_2 cell using Mid-IR Source.

The generated mid-IR source is invisible and has been checked using a broadband detector such as a mercury cadmium telluride (MCT) detector. In our case, we observed that the generated light operated in the mid-IR region using the absorption spectroscopy with the well-known transition of $^{12}CO_2$. For this work, we moved the pump wavelength from 857 nm to 850 nm, while the signal wavelength stayed at 1056 nm. Therefore, we expected that the idler wavelength was around 4.3 µm.

Figures 2 (a) and 2 (b) show the linear absorption signal at 4.3 μ m using a 170 mm long CO₂ cell with 1 Torr, and the expected absorption lines from the HITRAN database, respectively. The absorption strength and the spectral position of the signals were in good agreement with the expected values. From the results, the mid-IR source operated with a narrow bandwidth of less than Doppler broadening at the expected wavelength. In addition, from the absorption strength, the cavity enhancement technique was necessary for measuring ¹⁴CO₂ molecules.

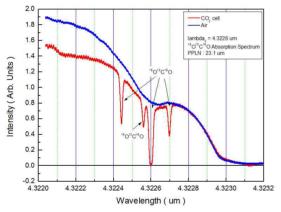


Fig.2(a). Absorption signals at around 4.3 μm by using mid-IR source.

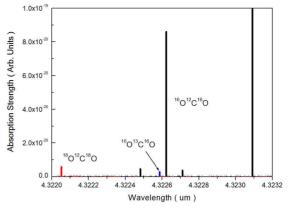


Fig. 2(b). The absorption strength and spectral position from HITRAN database at around 4.3 μ m.

3. Conclusions

We introduced a spectroscopic method to monitor $^{14}CO_2$ in gaseous effluent in a nuclear power plants to reduce the analysis time. As a first step, we demonstrated the mid-IR generation and tested the absorption spectroscopy using $^{12}CO_2$ molecules. From the results, the mid-IR source worked with a narrow bandwidth at the expected wavelength and the cavity enhancement technique was necessary for measuring the $^{14}CO_2$ molecules. We will provide a mid-IR source to the cavity enhancement technique for improving the sensitivity in the near future.

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