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Spectral Line Study for Measurement of Carbon-14 and H-3

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1. Introduction

Radiocarbon (C-14) and tritium (H-3) are the important radioactive isotopes utilized in environment monitoring around the nuclear facilities. These isotopes are low-energy beta emitters and present relatively low risk. However, prolonged exposure may affect living organisms. Therefore, continuous monitoring of these isotopes is required.

Currently, radiocarbon and tritium measurement employs the liquid scintillation counting (LSC) method. Despite its remarkable sensitivity, this technique shows disadvantages such as long sample collection time, intricate sample preparation and interference effects from background radionuclides. This, an alternative measurement technique is required to achieve on-line real-time detection of C-14 and H-3.

The optical measurement technique based on laser absorption spectroscopy offers direct detection of target molecules, in contrast to the conventional methods. It shows several advantages such as compact system size, cost-effectiveness, relative short measurement time, and usability. It is also free from interference effects. Based on these advantages, extensive research on the optically measuring radiocarbon [1-4] and tritium [5] has been conducted.

In this paper, we introduce and discuss about the spectral lines of $^{14}CO_2$ and HTO for trace-level detection of radiocarbon and tritium. Furthermore, we propose a specific wavelength range suitable for measurement for C-14 and H-3.

2. Methods and Results

In this section, we investigate the spectral line for radiocarbon and tritium measurement bases on spectrum databases [6, 7] and references papers. Then we present the wavelength range for measuring the C-14 and T-3.

2.1 Spectral Line Study for C-14 Measurement

The C-14 exists generally in the chemical form of carbon dioxide (CO_2) in atmosphere. Therefore, we consider the $^{14}CO_2$ as a main target for radiocarbon monitoring.

The carbon dioxide has the largest absorption band around 4.5 μm (about 2,200 cm^{-1}) wavelength region. Thus, this region is good candidate for the trace level $^{14}CO_2$ measurement. The previous studies [1-4] have performed in this wavelength range with differences in detailed absorption lines.

We can obtain the spectra of the various non-radioactive isotope compositions in the HITRAN database [6]. And the spectra of $^{14}CO_2$ which cannot extract form database, is obtained by theoretical calculation. Then, we suggested the optimal absorption line for $^{14}CO_2$ measurement by combining these spectra as shown in Fig.1.

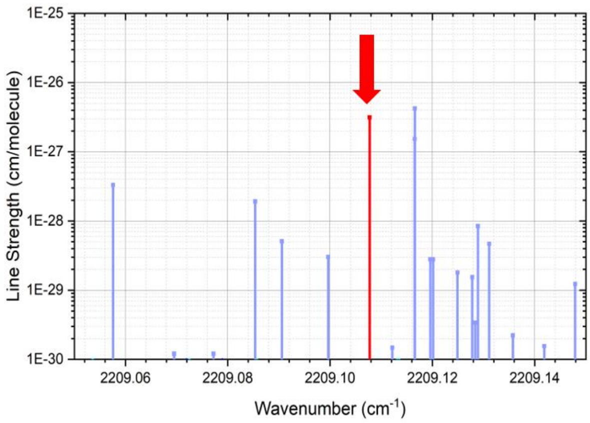

Fig. 1. Suggested absorption line for $^{14}CO_2$ measurement. This $^{14}CO_2$ concentration is 1 ppb in this spectra. The red lines represent the absorption lines of $^{14}CO_2$ and the light blue lines represent the absorption lines of CO_2 isotopologues except $^{14}CO_2$.

2.2 Spectral Line Study for H-3 Measurement

The tritium exists generally in the chemical form of water in atmosphere. Thus, we consider the HTO as the main target for tritium monitoring.

A previous work [5] reported a measurement sensitivity of 3 kBq (~1.3 MBq/g water) for optical measurement of HTO using a 2.17 μm wavelength range.

The spectra of the various isotope compositions of water can be extracted for the database in Ref. [7]. We can find what at the 4 μm wavelength range, HTO spectra show nearly 10 times larger absorption cross-section than that ate the 2.17 μm wavelength range. This suggests that the 4 μm wavelength region is a promising candidate for measuring trace level of tritium. To improve the measurement sensitivity, the absorption cross-section need to be maximized while isolating from other isotopologues.

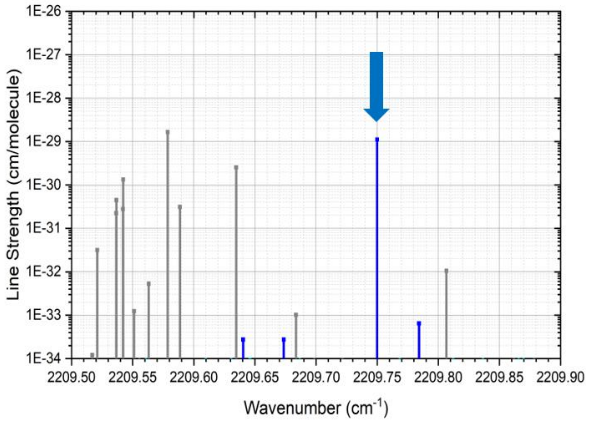

Fig. 2. Suggested absorption line for HTO measurement. The HTO concentration is 1 ppb in this spectra. The blue lines represent the absorption lines of HTO, and the gray lines represent the absorption lines of water isotopologues except HTO in natural abundance.

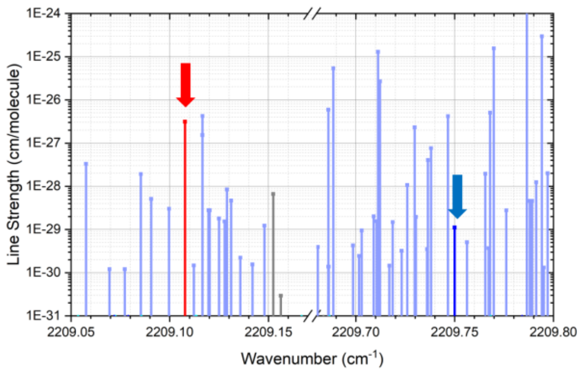

Fig. 3. Suggested absorption line for $^{14}CO_2$ and HTO measurement together. The $^{14}CO_2$ and HTO concentrations are 1 ppb each in this spectra. The red lines represent absorption lines of $^{14}CO_2$, the blue lines represent the absorption lines of HTO, the light blue lines represent the absorption lines of CO_2, and the gray lines represent the absorption lines of water.

We can find some possible lines for tritium monitoring by analyzing the spectrum data. The best line is shown in Fig. 2.

However, it is necessary to verify them experimentally, since these HTO spectra are extracted from calculation results. Depending on these results, these proposed lines can be changed.

2.3 Simultaneous Measurement of C-14 and H-3

The proposed wavelength ranges for HTO and $^{14}CO_2$ are in close proximity. This implies that if these lines fall within the wavelength tuning range of a single laser source, we can simultaneously measure these isotopes through a single instrument. Furthermore, we need not consider the contamination of equipment and sample preparation as the optical method is free from the interference by other radionuclides.

In addition, a simultaneous real-time measurement of radiocarbon and tritium may be possible in the absence of mutual interference effects. Fig. 3 shows the spectra of both CO_2 and water in the proposed wavelength region. For the $^{14}CO_2$ and HTO mixed sample, while the proposed line for $^{14}CO_2$ is still good candidate, HTO line is influenced by CO_2 isotopologues. This means that the CO_2 concentration of the sample affects the HTO measurement sensitivity.

3. Conclusions

We suggested the spectral lines for radiocarbon and tritium measurement in the mid-infrared range. In addition, we discussed about the possibility for their simultaneous measurement.

To achieve the proper sensitivity, we consider the spectroscopic techniques including a cavity ring-down spectroscopy (CRDS). In the future, we will build experimental devices and measure the $^{14}CO_2$ and HTO in the proposed wavelength ranges.

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