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Heavy Water Concentration Measurement in Air

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1. Introduction

The PHWRs are the backbone of Indian nuclear program and heavy water is one of the key input materials in PHWRs. Since, in the coolant channels, heavy water flows at high temperature and pressure; leaks in the heat transport system are not uncommon. The loss of heavy water due to such leaks can lead to spreading of radioactivity and it also contributes to operating cost of the nuclear reactor. Furthermore, it is advantageous to detect small leaks, because if remains undetected, they may develop into a severe leak, which may lead to reactor shutdown. In view this, it is important to detect minor leaks of heavy water in PHWRs [1].

There are many techniques are available to detect leakages of heavy water in nuclear reactors. Such as Beetle detectors, dew point sensors and tritium activity monitors. The beetle detectors are placed under susceptible joints and detect the leaks by shorting of electrodes due to presence of heavy water. Beetle detectors cannot detect minor leaks due to evaporation of heavy water, since coolant in the pressure tube flows at a high temperature and pressure. Dew point sensors, monitors the heavy water leak from primary coolant to annulus gas inside the reactor core. The tritium activity monitoring is normally used to monitor heavy water leaks to atmosphere and based on tritium beta activity. However, this method is indirect and time consuming. In view of above limitations of present leak monitoring system, we have developed a trace HDO detector using cavity enhanced laser absorption spectroscopic technique [2,3]. This method directly determines the concentration of HDO in air and water samples. Since, heavy water readily reacts with water to form HDO as per following reaction

$$H_2O + D_2O \leftrightarrow 2HDO \qquad (1)$$

The concentration of D_2O in sample is half the HDO concentration (applicable only for small D/H ratio in sample). The details of the developed trace HDO detection setup have been presented here.

2. Experimental

The experimental setup used for HDO concentration measurement in air using OA-ICOS technique is shown in Figure 1. The optical cavity consists of 2 inch diameter high-reflectivity plano-concave mirrors, with specified reflectivity of 99.98% at 1390 nm and radius of curvature 1 m placed 14.5 cm apart. The inlet and outlet ports are provided on the flanges and the flow rate at both ports are controlled by bellow sealed valves, which are used for flow control. The outlet port is connected to a turbo-molecular pump. A discrete mode (DM) diode laser with TEC controller from Eblana photonics with a centre wavelength of 1390.5 nm has been used as a source. A fibre coupled laser beam is collimated and directed to the cavity without mode matching. The laser and the optical cavity are aligned to maximize the signal to noise (S/N) ratio. The laser wavelength is ramped by applying a saw tooth wave with an amplitude 0.12V and a 0.93V offset at a 100 Hz rate to the Vbias input of the laser module. This scans the laser frequency from 7190.7-7191.6 cm-1. The transmitted light is focused on a germanium photodiode, with a pair of lenses. The transmission signal of the cavity and the ramp voltage are recorded on a digital oscilloscope. The signal is averaged for 100 cycles. The transmission signal and the ramp voltage are then processed to obtain the absorption spectrum and concentration of HDO in sample is calculated by suitable curve fitting.

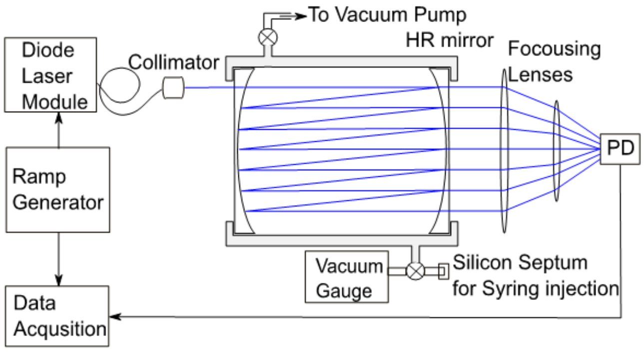


Figure 1 Schematic of the experimental setup. HR: high reflectivity; PD: photodiode.

3. Results and Discussion

The HDO concentration in the laboratory has been measured. At the time of measurement the temperature of the laboratory was 25.5°C with 50±5% relative humidity. This corresponds to fraction of water vapor in air of 1.43 to 1.75 %. The isotopic composition of water vapor near the sea surface and in coastal areas (like Mumbai) is deficient in D/H ratio by 70 to 180 mil. Hence the HDO concentration in the laboratory air is 4.12 to 4.67 ppm. For such small concentration measurements with sufficient SNR, the pressure inside the cell has to be optimized. This optimization is carried out by estimating the height to width ratio of cavity absorption signal as shown in Figure 2. Maximum height to width ratio of HDO absorption line is obtained at 100 mbar.

The laboratory air is sampled continuously using flow through system. The pressure inside the cell is maintained at 100 mbar by controlling the inlet and outlet valves. The absolute H216O and HDO concentrations are obtained using area under the single pass absorption signal using Beer-Lambert law [4] and are shown in Figure 3(a). For estimation of the error in measurement, a large number of absorption signals have been acquired and processed. The measured average H216O concentration is 1.54±0.07% which is within the specified range. The measured HDO concentration is 4.27±0.38 ppm, is also within the range. The sensitivity of each measurement is thus 0.38 ppm and sensitivity of 0.1 ppm is achieved with 20 averaging.

In the above measurements the sensitivity is mainly limited by the intensity noise due to optical interference effects in the cavity and other optical components. The effect of these can be minimized by dithering the cavity length. The effect of dithering is to average out the mode structure. The cavity length dithering is provided by placing a CPU fan close to the cavity. The absorption spectrum thus obtained is shown in Figure 3(b). Figure shows considerable improvement in signal to noise ratio, due to averaging out of cavity mode structure. The sensitivity of 45 ppb has been achieved with 20 averaging.

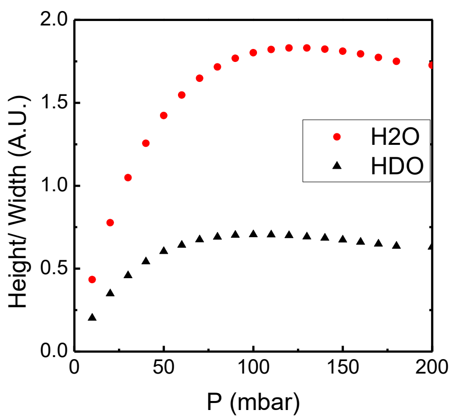
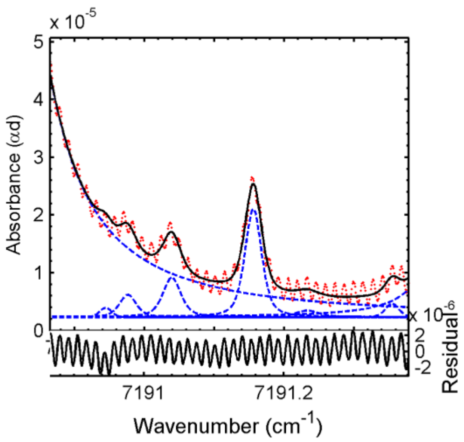
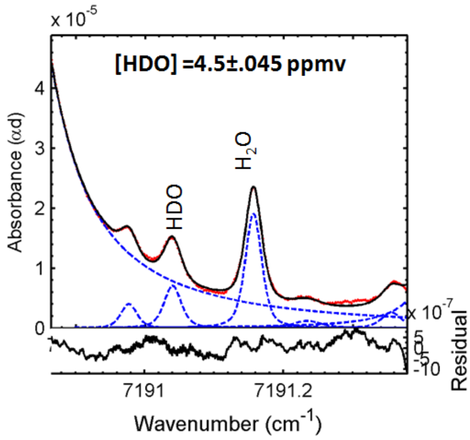


Figure 2 Optimization of pressure



(b)



(c)

Figure 3 Measurement of HDO concentrations in laboratory air: (a) Observed spectrum without cavity length dithering; (b) Observed spectrum with cavity length dithering.

4. Conclusion:

An experimental setup based on OA-ICOS technique has been demonstrated for the measurement of heavy water (HDO) concentration in laboratory air. In this technique the mode noise associated with the optical cavity is the major source of noise. This has been successfully overcome by using cavity length dithering. The demonstrated setup has achieved the sensitivity of 45 ppb with averaging. This sensitivity in HDO concentration measurement in air is sufficient for the application in PHWR heavy water leak detection system.

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