Jinst

Published by IOP Publishing for Sissa Medialab

Received: *November 14, 2019*
Accepted: *February 20, 2020*
Published: *March 20, 2020*

Laser based technique for monitoring heavy water leaks in nuclear reactors: performance validation with conventional techniques

A. Gupta,[a,b] **S. Prasad,**[c] **S.A. Prabhu,**[d] **S.P. Somkuwar,**[a] **K.P. Younus,**[c] **S. Raghunathan,**[a] **D.V. Udupa,**[a,b] **M.N. Deo**[a,b] **and A. Topkar**[b,e,1]

[a] *Atomic and Molecular Physics Division, Bhabha Atomic Research Centre, Mumbai, 400085, India*
[b] *Homi Bhabha National Institute, Mumbai, 400094, India*
[c] *Radiation Safety Systems Division, Bhabha Atomic Research Centre, Mumbai, 400085, India*
[d] *Heavy Water Division, Bhabha Atomic Research Centre, Mumbai, 400085, India*
[e] *Electronics Division, Bhabha Atomic Research Centre, Mumbai, 400085, India*

E-mail: anita@barc.gov.in

Abstract: In a pressurized heavy water reactor (PHWR), monitoring leakages of heavy water from coolant and moderator systems is very important for reactor safety as well as health of operating personnel. In view of this, heavy water leak monitoring system based on off-axis integrated cavity output spectroscopy (OA-ICOS) technique is developed. The developed system estimates the amount of heavy water in the samples by measuring the HDO concentration with the detection sensitivity of ±1.2 ppm. The performance of this system is studied by measuring various samples obtained from the Dhruva research reactor at Bhabha Atomic Research Centre. The validation of the data obtained using this system is carried out using tritium activity measurements of same samples. The results are also compared with D/H ratio measurements obtained by isotope ratio mass spectrometer (IRMS). The measured HDO concentration of various samples using the developed system shows a good correlation with the measurements using IRMS and tritium activity measurement techniques.

Keywords: Spectrometers; Lasers

[1] Corresponding author.

https://doi.org/10.1088/1748-0221/15/03/P03018

2020 JINST 15 P03018

Contents

2020 JINST 15 P03018

1 Introduction

In Pressurised Heavy Water Reactors (PHWRs), heavy water is used both as coolant and moderator. Moderator is required to slow down neutrons effectively for a sustained chain reaction. During the normal operation, heavy water is contained in two separate closed loop systems each for moderator and coolant [1]. Although, nuclear reactors implement various safety features for containment, small amount of leakages of heavy water (coolant as well as moderator) still occur during normal reactor operation. These leakages are mainly through seals, valves, pipelines and steam generator, etc. Some heavy water leakages also occur during refueling through feeders, end-fittings and the closure plug seals. This loss of heavy water due to leakages requires makeup to compensate for the escapes. Portion of leaked heavy water is usually recovered; since it is usually downgraded, it requires cleanup and up-gradation [2]. This adds to the cost of reactor operation [3].

Besides the normal loss of heavy water, there could be some additional leakages because of degradation or malfunctioning of reactor system due to corrosion or cracking in the reactor components. Such leak events may lead to reactor shutdown and/or release of radioactivity to the environment. Radiation hazards are primarily due to the activation of water molecules and dissolved gases by high neutron flux in the reactor. The induced activity in water generates the isotopes ^{16}N, ^{17}N, ^{19}O and ^{3}H [4]. The radio-nuclides, ^{16}N, ^{17}N and ^{19}O produce an intense gamma and beta radiation, limiting the access to the equipment in a running reactor. Due to short half-life, these three isotopes decay fast after a shutdown. However, the tritium isotope is a low energy β emitter with a half-life of 12.3 years. This long half-life leads to its concentration build-up in heavy water systems, which hardly decrease on shut down. Thus a leak in heavy water system during normal operations and accidents can produce dangerous levels of tritium in the atmosphere. Though, the low energy beta particle from tritium is not an external radiation hazard, it can do internal damages if it enters the body through inhalation and skin [5].

Since the leaks in heavy water system are indication of reactor health, a continuous heavy water leak monitoring system is essential to avoid any major leak situation. A few techniques are available to detect leakages of heavy water in nuclear reactors such as using Beetle detectors,

dew point sensors, ^{19}O monitors and tritium activity monitors [6, 7]. All these techniques have some advantages as well as some limitations over the others. For example, the beetle detectors are the simplest available heavy water leak detectors which are placed under susceptible joints. They detect the leaks due to shorting of electrodes as a result of accumulation of leaking coolant (or moderator) into the collection pots. However, they cannot detect minor leaks due to instantaneous evaporation of leaking coolant, on account of high temperature and pressure inside the pressure tubes. Similarly, the use of dew point sensors is limited to monitoring of coolant leaking from the primary coolant channel to the annulus gas inside the reactor core. In addition, both these sensors cannot distinguish between H_2O and D_2O which may lead to triggering of false alarm [8]. The online ^{19}O monitors are used for detecting leakage of primary coolant to secondary coolant in heat exchangers by monitoring the 1.357 MeV γ-photons of ^{19}O [9]. The sensitivity of these detectors is limited due to the interference caused by other radio nuclides such as ^{41}Ar, ^{85}Kr, ^{222}Rn or ^{133}Xe emitting γ-photons (energy ranging from 81 to 1294 keV) and β-particles (energy ranging from 346 to 1197 keV) in the reactor environment. Mostly, monitoring of heavy water leak to the atmosphere is done by tritium beta activity monitoring using a Liquid Scintillation Counter (LSC) [10]. The liquid samples obtained by condensing the air at various locations are collected manually and their activity is measured off-line. For the measurement of tritium in air, either bubblers or cold fingers are used to collect the moisture in the air. This involves a delay due to the time required for sample collection, preparation and counting.

None of the methods discussed above meet all the requirements at the same time viz. high sensitivity, high selectivity, real time online measurement capability, portability, high reliability, and ease of use. Thus there is strong need to develop a monitoring system which meets the maximum requirements. For the measurement of heavy water leakage in a nuclear reactor, it is desirable to use monitoring systems which are based on entirely distinct processes. The advent of compact tunable diode lasers in near IR range has revolutionised the development of portable gas detectors [11–14]. Hence, LAS technique for the detection HDO is selected in the present work, since the concentration of HDO is related to the concentration of heavy water in air and water. A major merit of LAS based system for coolant leak detection is the selectivity and negligible interference from the other radioactive species inherently present in the reactor environment. In addition, the laser based technique is suitable for in situ online real time measurement of the HDO concentration without a need for any lengthy sample preparation.

We have developed a trace HDO detection system for monitoring heavy water leaks in PHWRs. The details of this system and calibration results are presented earlier [15]. The system is based on an advanced LAS technique known as off-axis integrated cavity output spectroscopy (OA-ICOS). The technique basically utilizes an optical cavity for enhancing the interaction path length of laser beam with sample for enhancing the absorption signal [16–18]. In this paper, the stability analysis and the validation of the system with conventionally used techniques is presented. The results of measurement on samples collected from Dhruva research reactor demonstrating the performance of the system are discussed. A comparison of results with other established techniques like tritium monitoring and Isotope Ratio Mass Spectrometer (IRMS) is presented.

2020 JINST 15 P03018

2 System description

The heavy water leak monitoring system for PHWR presented in this paper is based on measurement of HDO concentration in surrounding air or secondary (light water) cooling system. The change in the concentration of HDO in the surroundings is proportional to the amount of leaked heavy water, since D_2O readily reacts with water (H_2O) to form HDO. The details of the developed OA-ICOS based heavy water monitoring system are described earlier and are briefly presented in this paper [15]. Figure 1 shows the photograph of the developed system. The system comprises of optical cavity, lasers, detectors, and vacuum components. The main specifications of various subsystems are listed in table 1.

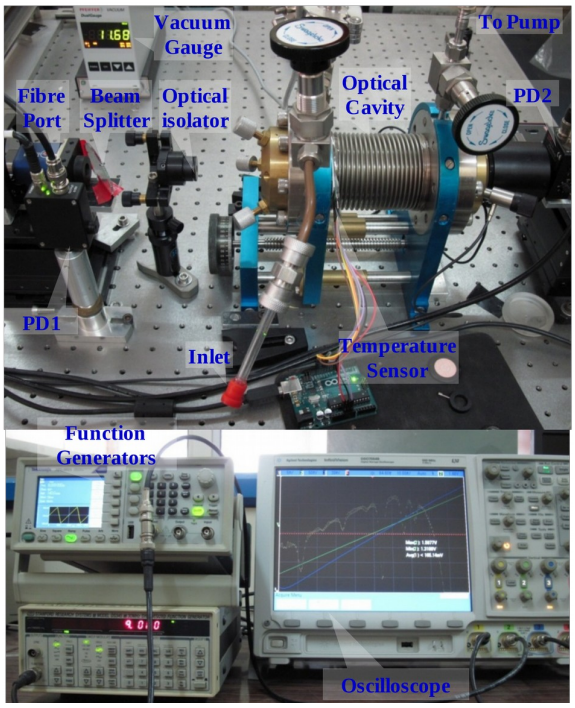

Figure 1. Photograph of the developed heavy water detection system. (PD1 and PD2: germanium photodiodes 1 and 2, T: Temperature sensor).

The cavity output is related to the wavelength dependent absorption coefficient, $\alpha(\lambda)$, by the following equation:

$$I(\alpha) = I_{\text{in}} C_p T^2 \frac{e^{-\alpha(\lambda)d}}{1 - \left(Re^{-\alpha(\lambda)d}\right)^2} \tag{2.1}$$

here I_{in} is incident laser intensity, C_p is coupling efficiency, R is mirror reflectivity, T is mirror transmittance and d is distance between the mirrors. The absorption spectra, $\alpha(\lambda)$ of the sample is obtained by solving this equation.

For recording the cavity transmission signal, the laser wavelength is scanned across HDO and $H_2^{16}O$ lines at 1390.6196 and 1390.5968 nm respectively by linearly varying the laser diode current. The output of both photo-diodes and the wavelength scanning ramp signal were digitized using an oscilloscope. The absorption spectrum, that is absorbance per pass versus wavenumber,

2020 JINST 15 P03018

Table 1. Specifications of various sub-systems.

S. No.	Description	Specification
1	High reflectivity mirrors	Aperture: 2 inches
		ROC: 1 meter
		Reflectivity: 99.92%
2	Distance between the mirrors	13.5 cm
3	Effective path length	168.75 m
4	IR Source: Discrete mode diode laser	Centre wavelength: 1390.6 nm
		Tuning range: ±1 nm
5	Detectors: Germanium photo-diodes	Sensor diameter: 5 mm
		Wavelength range: 800–1800 nm
		Switchable Gain
6	Vacuum Gauge: Capacitance manometer	Range: 0.1 to 1100 hPa
		Accuracy: 0.2% of reading
7	Vacuum pump	Turbo molecular pump
8	Temperature sensor, TSYS01	Accuracy: ±0.1°C,
		Temperature resolution: 0.01°C
9	Beam splitter: Pellicle beam splitter	Splitting ratio: 8:92

is calculated from the acquired signals using equation (2.1). The concentration of HDO and $H_2^{16}O$ molecules is computed from the area under the respective absorption peaks and known spectral constants (HITRAN database [19]).

3 Performance evaluation

The performance of the developed OA-ICOS based system is studied by measuring the concentration of HDO in the water samples collected from different locations in Dhruva research reactor, at Bhabha Atomic Research Centre. These samples include secondary cooling water sample and bubbler samples at certain locations in the reactor. The performance is validated by comparing the results with those obtained by conventional techniques i.e. LSC and IRMS.

The samples were injected in the optical cell using a syringe. For each measurement, the syringe and the cell were cleaned thoroughly with the sample itself to remove any residue of previous sample. The cell is evacuated to a pressure of 10^{-3} mbar. Four µlitres of water sample is then injected into the cell through the silicon septum, creating a sample vapour pressure of approximately 12 mbar inside the cell. The resultant optical transmission signal along with the pressure and temperature of cell were measured and recorded. The HDO concentration is determined for each sample after processing the obtained signal and by curve fitting the spectrum as described in earlier report [15].

The sensitivity for concentration measurement by the laser spectroscopic method can be improved significantly by averaging. However, the averaging time is limited by the time dependent drifts in the system. Thus it is important to quantify the maximum averaging time within which the sensitivity of the detection system can be improved by means of averaging. The maximum averaging time for the developed system is estimated by means of Allan Variance. For the determination of

2020 JINST 15 P03018

maximum averaging time, the HDO concentration a sample is recorded for whole day. The data is processed to determine HDO concentration and the time series of the HDO concentration is recorded. The time series plots of recorded HDO concentration is shown in figure 2(a). The Allan variance plot (figure 2 (b)), for the same sets, shows that optimum averaging time for measurement is 1280 sec. It can also be noted that the standard deviation of less than ±1.5 ppm can be obtained by averaging for 160 sec (figure 2 (b)). This sensitivity can be considered sufficient for online monitoring of heavy water leaks in PHWRs.

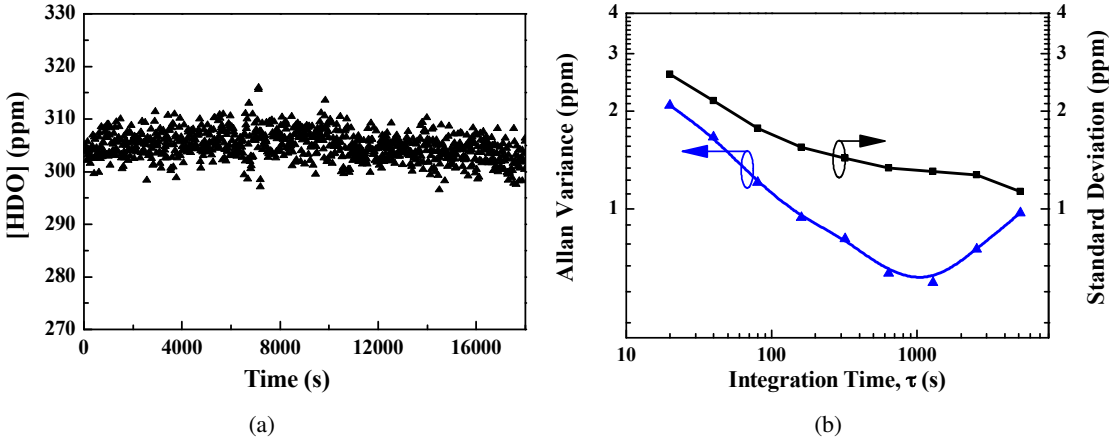

Figure 2. (a) Time series plot of recorded HDO concentration; (b) Allan variance and standard deviation plot.

For measurement of HDO concentration in various samples, an average value of measurements for 1280 seconds is used for determining the HDO concentration. Figure 3 shows the plot of such measurements for seven different samples, labeled S1 to S7. The sample labeled S1 is the standard distilled water, for which the measured value of HDO concentration is 300.6 ±1.74 ppm. The measured average value along with the standard deviation for other samples is also shown in the table 2. The HDO concentration in all the samples is found to be higher than that of normal water indicating leaks of heavy water (D_2O) from the heat transport system of the nuclear reactor.

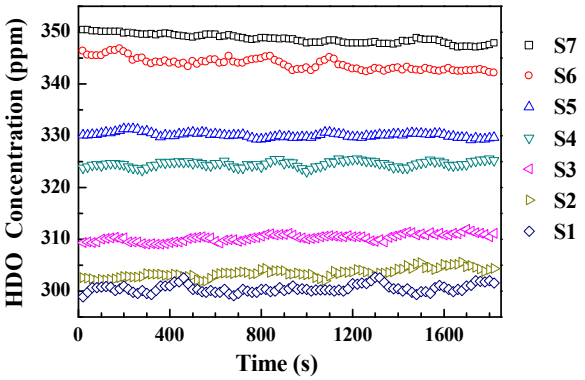

Figure 3. A plot showing continuous measurement of HDO concentration in samples labeled S1 to S7. The measured average value of HDO concentration in ppm and standard deviation for each sample is given in table 2.

2020 JINST 15 P03018

Table 2. Measured average HDO concentration in various samples for averaging time of 1280 seconds.

Sample Label	Measured Average Value of HDO Concentration in ppm	Measured Standard Deviation
S1	300.6	1.74
S2	303.6	2.18
S3	310.4	1.56
S4	324.6	1.61
S5	330.1	1.12
S6	343.6	2.21
S7	348.5	1.17

In order to validate the performance of the system for actual deployment in a nuclear reactor, the correlation between the measured HDO concentrations with the measurements obtained using other established techniques is necessary. As discussed earlier, the alternate technique used to detect heavy water leak in a nuclear reactor is based on tritium monitoring. For isotope ratio measurement of hydrogen, traditionally IRMS is used. Hence, the performance validation of the developed system is based on the correlation of HDO concentration measurements with the data obtained using these techniques:

3.1 Correlation with tritium monitoring

Tritium monitoring is the most sensitive method available and hence is being used in PHWRs worldwide for monitoring heavy water leaks in the heat transport system. A high level of tritium is formed due to neutron activation of deuterium in the coolant and moderator systems. Any leak in these systems will result in rise in both deuterium and tritium concentrations in the surrounding light water system (e.g. secondary coolant) and in the surrounding environment. Since both the systems contain high purity heavy water, the changes in deuterium concentration and tritium concentration in the surrounding light water samples are linearly correlated. The proportionality constant between these two isotopes depends on the activity of coolant and moderator. Thus, the tritium monitoring system is an indirect method, which is being used to estimate the losses of heavy water occurring to the surroundings.

In the present work, the tritium concentration (T_β) for all the samples is measured by a LSC. The tritium concentration in the samples is compared with the change in deuterium concentration (δD) in the samples. Here, δD (in mils) is obtained by comparing the HDO concentration of a standard sample, using the following equation:

$$\delta D = \frac{HDO_{sample} - HDO_{standard}}{HDO_{standard}} * 1000 \tag{3.1}$$

The plot of measured δD versus normalized tritium concentration is shown in figure 4. Each sample is measured three times, and the average value is obtained. The HDO concentration and the T_β were expected to follow the same proportionality for all samples. The measured δD shows a linear variation with tritium concentration. The obtained correlation coefficient between T_β and δD is 0.9519. The data presented in figure 4 confirms that the sensitivity of the developed system for

2020 JINST 15 P03018

heavy water leak monitoring is comparable to that of a tritium monitoring system based on liquid scintillation counting.

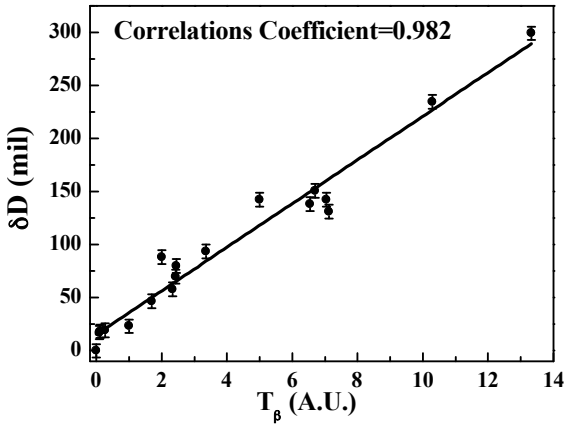

Figure 4. Comparison of measured change in deuterium concentration δD using developed OA-ICOS system with tritium concentration measured using LSC.

Due to routine leaks in the heat transport system, there is a measurable level of T_β and δD in the reactor system. Additional leaks in the reactor system are indicated by an accompanied rise in both of them. Such leaks manifests wear and tear in the system which may develop into severe leaks if remain unattended. For the safety of the reactor and people working there, it is important to locate the leaks at the earliest. Detecting a leak location in PHWRs is a tedious and time consuming task. Since, heavy water is used both as coolant and moderator in two isolated systems; the leaky system can be distinguished based on the difference in tritium concentration. The difference in tritium concentrations in the coolant and moderator channels is due to difference in their volume and residence time inside the reactor core. Therefore, if the ratio of tritium to heavy water in the sample is known, it can be used to identify the system from which heavy water is leaking [20]. Thus, HDO concentration measurement system along with the tritium monitors will be useful not only in determining heavy water losses but also in the diagnosis of a faulty leaking system.

3.2 Correlation with IRMS

IRMS is one the most sensitive techniques to measure isotopic ratios of H, C and O which finds application in environmental science [21]. However, it is not suitable for leak monitoring system in PHWR due to its size, cost, sample preparation, and handling considerations. Moreover it cannot be applied for online monitoring. In this study, we have compared the results of HDO concentration measurements by the developed system with D/H measurements by IRMS to cross-check the calibration and to investigate whether the measurement sensitivity is comparable to that of IRMS.

The D/H ratio of all the samples obtained from Dhruva reactor is also measured with available IRMS equipment with sensitivity of ±1 ppm. Figure 5 shows the plot of correlation between HDO concentration by OA-ICOS and D/H ratio measurement by IRMS. The correlation coefficient of OA-ICOS measurements and IRMS measurement is 0.9766. The measurement sensitivity of the

2020 JINST 15 P03018

developed system is ±1.2 ppm for HDO concentration in water, which is equivalent the sensitivity of D/H ratio by the IRMS instrument.

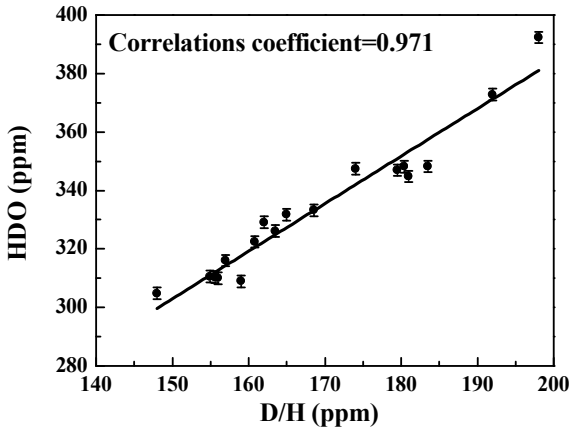

Figure 5. Correlation between HDO concentration by OA-ICOS and D/H ratio measurement by IRMS.

4 Conclusion

An OA-ICOS based system aimed for monitoring heavy water leaks in PHWRs is developed. The system is analyzed for stability and maximum averaging time is evaluated to be 1280 seconds. The sensitivity of developed OA-ICOS system is observed to be 1.2 ppm. In addition, it has been established that 1.5 ppm sensitivity can be obtained with 160 sec averaging, which is sufficient for online monitoring of heavy water leaks in PHWRs. Lee et al. have earlier demonstrated the detection of small D_2O leak from a fuel channel closure plug in Wolsong-4 PHWR [22, 23]. The measurements were done using the samples only at one location in the reactor and the comparison of their results with conventional techniques used for heavy water leak monitoring was not presented. In this paper, the cross-correlations between the measurements of HDO concentration by the developed system with the data obtained using the existing established techniques are presented. The technique is validated by measuring the samples obtained from different places in the Dhruva reactor. The samples were also analyzed by conventional techniques viz. LSC based tritium monitors and IRMS. All three techniques show a very good correlation. A good correlation between analysis by OA-ICOS system and LSC indicates that the sensitivity of the developed OA-ICOS based is as good as LSC. The sensitivity of the present system has been found to be comparable with the IRMS. The advantages of developed OA-ICOS system over conventional tritium or ^{19}O monitoring techniques would be possibility of online real time interference free monitoring and portability. The system will be further improvised for HDO concentration measurement in air.

2020 JINST 15 P03018

References

[1] S.S. Bajaj and A.R. Gore, *The Indian PHWR*, *Nucl. Eng. Des.* **236** (2006) 701.

[2] T. Park and K. Aydogdu, *Effect of design improvements in heavy water management systems to reduce heavy water losses and tritium releases at Wolsong 2,3 and 4*, International Atomic Energy Agency (IAEA) (1994), pp. 1011 – 4289.

[3] M.V. Ramana, D.S. Antonette and K.N.R. Amulya, *Economics of Nuclear Power from Heavy Water Reactors*, *Econ. Polit. Wkly* **40** (2005) 1763.

[4] AERB, *AERB Safety Guide- Radiation protection aspects in design for pressurised heavy water reactor based nuclear power plants*, (2005).

[5] D. Delacroix, J.P. Guerre, P. Leblanc and C. Hickman, *Radionuclide and radiation protection data handbook 2002*, second edition, Nuclear Technology Publishing, (2002).

[6] A.I. Miller, J. Luxat, E.G. Price and P.J. Fehrenbach, *Heavy water reactors*, in *Nuclear Engineering Handbook*, second edition, CRC Press, (2016), pp. 163–220.

[7] T.-K. Park and S.-H. Jung, *Detection of gaseous heavy water leakage points in CANDU 6 pressurized heavy water reactors*, *INIS*, vol. 30, (1996).

[8] B.B. Zhai, *Systems and methods for detecting a leaking fuel channel in a nuclear reactor*, Google Patents, (2017).

[9] S.R. Mitra et al., *Enhancement of efficacy of process water monitors in detecting heavy water leak in steam generator blow down lines*, Prime Time Education, India, (2006).

[10] B.N. Dileep et al., *Counting of samples labeled with tritium using binary liquid scintillation solution*, *Radiat. Protect. Environ.* **24** (2001) 152.

[11] R.F. Curl and F.K. Tittel, *Tunable infrared laser spectroscopy*, *Annu. Rep. Prog. Chem.* **C 98** (2002) 219.

[12] E. Kerstel and L. Gainfrani, *Advances in laser-based isotope ratio measurements: selected applications*, *Appl. Phys.* **B 92** (2008) 439.

[13] M. Sigrist, R. Bartlome, D. Marinov, J. Rey, D. Vogler and H. Wächter, *Trace gas monitoring with infrared laser-based detection schemes*, *Appl. Phys.* **B 90** (2008) 289.

[14] M. Lackner, *Tunable diode laser absorption spectroscopy (TDLAS) in the process industries–a review*, *Rev. Chem. Eng.* **23** (2007) 65.

[15] A. Gupta, P.J. Singh, D.Y. Gaikwad, D.V. Udupa, A. Topkar and N.K. Sahoo, *Instrumentation and signal processing for the detection of heavy water using off axis–integrated cavity output spectroscopy technique*, *Rev. Sci. Instrum.* **89** (2018) 023110.

[16] R. Daniele, I. Ventrillard, G. Méjean, J. Morville and E. Kerstel, *Introduction to Cavity Enhanced Absorption Spectroscopy*, in *Cavity Enhanced Spectroscopy and Sensing*, G. Gagliardi and H.-P. Loock, Eds., Springer, London, U.K. (2014).

[17] A. O'Keefe, J.J. Scherer and J.B. Paul, *cw Integrated cavity output spectroscopy*, *Chem. Phys. Lett.* **307** (1999) 343.

[18] V.L. Kasyutich, C.E. Canosa-Mas, C. Pfrang, S. Vaughan and R.P. Wayne, *Off-axis continuous-wave cavity-enhanced absorption spectroscopy of narrow-band and broadband absorbers using red diode lasers*, *Appl. Phys.* **B 75** (2002) 755.

2020 JINST 15 P03018

[19] L.S. Rothman et al., *The HITRAN 2008 molecular spectroscopic database*, *J. Quant. Spectrosc. Radiat. Trans.* **110** (2009) 533.

[20] J. Shin, *A Method of Locating Leakage of Heavy Water in CANDU Reactors Using Ratio of Tritium to Heavy Water*, presented at the ISOE Asian ALARA Symposium, Aomori, Japan, (2009).

[21] R.K. Boyd, C. Basic and R.A. Bethem, *Trace quantitative analysis by mass spectrometry*, John Wiley & Sons, (2011), [DOI].

[22] H. Park et al., *A laser leak detector for leakage monitoring of fuel channel closure plugs in pressurized heavy water reactors*, *Nucl. Eng. Des.* **265** (2013) 880.

[23] L. Lee, H. Park, T.-S. Kim, M. Kim and D.-Y. Jeong, *Development of a portable heavy-water leak sensor based on laser absorption spectroscopy*, *Annals Nucl. Energy* **87** (2016) 350.

2020 JINST 15 P03018