# A Real-time Tritium–In–Water Monitor for Measurement Of Heavy Water Leak To The Secondary Coolant

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## 1. Introduction:

In a pressurized heavy water reactor (PHWR), large inventory of high purity heavy water is needed as it is used both as primary coolant and moderator. In this type of reactor light water is employed as secondary coolant. In the fresh heavy water tritium activity is negligible. In the reactor because of very high neutron flux, the deuterium in the heavy water is converted tritium by neutron capture. Therefore, with the passage of time, tritium activity concentration in the heavy water goes on increasing. Since the primary coolant (heavy water) being at high temperature and pressure, the instances of heavy water leakage to the secondary coolant are not uncommon. Though moderator is at lower temperature and pressure, leakage of moderator also takes place. Leakage of tritium is also a major radiation hazard in PHWRs. The high cost of heavy water demands that its loss should be prevented to the maximum extent possible. This measure will also ensure normal and healthy functioning of the plant. Therefore, it is important to develop method and techniques which will detect and measure the quantity of heavy water leakage. This requirement has necessitated the use of continuous on-line monitoring system to monitor the leakage of heavy water to the secondary coolant. It is important to note that once the leakage is detected it is much easier to control it.

At present the system used in the reactors is based on measurement of the quantity of tritium by liquid scintillator spectrometer. The sample is collected manually and its activity measured by using this system. This involves delay due to sample collection, preparation and counting. Though the use of an on–line liquid scintillator system is described in literature, not a single system is commercially available(1).

Solid scintillator system based on the plastic scintillator film packed flow cell was developed at BARC(2). This system is very useful for on–line continuous monitoring of tritium in the secondary coolant line. A proto type model is in operation at Rajasthan Atomic Power Station for quite some time(3). Based on the operational experience some modification are carried out in the unit.

## 2. Development Of tritium In Water Monitor At BARC:

The monitor consists of three parts. (i) Detector assembly, (ii) Sample conditioner, and (iii) Electronic system. Fig.1 shows the schematic of tritium in water monitor with sample conditioner.

### 2.1 Detector Assembly:

A very thin 5 μm thick and 500 $cm^2$ area plastic film scintillator has been developed at BARC, for tritium measurement. Since the range tritium beta of in water is around 0.005 mm, beta particles which are in contact with the scintillator, can only be detected. To have a better sensitivity, the detector should have a large surface area. By Packing 6 films in the flow cell one can obtain a surface area of approximately 3000 $cm^2$. This scintillator film which has large number of small holes, when packed in the flow cell, make it highly porous and behaves like a sponge thus allowing a smooth flow of the sample. Two EMI-9635 photomultiplier tubes are optically coupled on either face of the flow cell and measurement is done in coincidence mode. Sample water is passed continuously through the flow cell. Scintillations (light photons), produced in the detector are proportional to the activity present in the flow cell. Intensities of these photons is measured using the P.M. tubes and subsequent electronics. Fig. 2 shows the tritium activity concentration versus the count rate with 6 numbers of films packed in the flow cell.

### 2.2 Sample Conditioner:

When secondary coolant water is passed through the flow cell, impurities and particulate substance in the sample water get trapped in the plastic sponge detector. This results in reduced transparency of the detector, thus affecting sensitivity. It was observed that the chlorine which is added in the secondary coolant to arrest the

fungus formation gives chemiluminescence, which gives an erroneous signal. In addition to this, it was also observed that the dissolved radioactive argon present in the sample water also interferes with the measurement.

To overcome the various problems such as reduction in the detector sensitivity, interference due to chemiluminescence and the dissolved radioactive gases, a special type of sample conditioner is designed. The sample conditioner consists of water filtration unit granular type activated charcoal and a bubbler system.

Water filter consists of polypropylene bag in which a polypropylene candle of 5 μm pore size filled with granular type activated charcoal and filter paper assembly consisting of glass paper and Whatman filter papers are put. Except filter paper assembly, all other parts are commercially available and are easy to replace. Polypropylene bag removes turbidity in the sample, polypropylene candle traps particulates above 5 μm size and activated charcoal absorbs organic compounds, free chlorine, fungus and turbidity. Filter papers are used for fine filtration. To filter submicron size particle, glass fibre filter paper sandwitched between Whatman filter paper 541 and 540 is used.

For removing the gaseous radionuclide interference in the sample water, a bubbler system is used. Non active gas such as nitrogen is bubbled through a sintered disc which strips out the dissolved radioactive gases from the sample water. The bubbler is found to be very effective in removing the dissolved radionuclide interference.

## 2.3 Electronic System:

Electronic system consists of a high voltage unit, coincidence amplifier, anti-coincidence analyzer, microprocessor based digital counter, alarm system, printer and 4 – 20 mA output to the control room. Fig. 3 shows the block diagram of the electronics system. High voltage unit provides the accelerating potential to the photomultiplier tube. Main part of the background counts is due to thermal noise in the P.M. tubes. Since it is random in nature, its contribution can be substantially eliminated by using coincidence circuitry. The anti-coincidence analyzer provides energy discrimination against high-energy radiation and enables counting in the window corresponding to tritium beta. The microprocessor system enables the selection of counting time and provides two separate channels one for tritium betas and the other for the particles of betas high energy which is actually an interference. Two channel counting enables to know whether there is any interfering radiation present and for enabling the compensation against interference. By selection of higher counting times one could achieve better sensitivity. Alarm level can be fixed as per the user's requirement. The tritium activity value in the sample is printed using the printer.

## 3. Discussion:

Tritium in water monitor with sample conditioner is tested at laboratory for finding the suitability of the sample conditioner. First, the background measurement is carried out. Then the detector sensitivity is found out by passing 100 nCi/ml of tritium activity through the detector. Tap water is connected at the inlet of the system as shown in Fig.1. Water flow rate is adjusted to around 100 ml/minute through the flow cell detector. Detector sensitivity is periodically checked by passing varying quantity of tap water through the detector. Filter paper is replaced when it gets blocked. This happens approximately after one week of continuous use. The polypropylene candle filled with charcoal is replaced after a month. Fig .4 shows the plot of sensitivity versus the quantity of water passed through the detector. After passing around 2200 litres of tap water sensitivity falls by about 12% from its original value. By this rate if the water quality is assumed to be same, may need around 8000 litres of tap water flow through the detector for a 50% fall in sensitivity. A sensitivity fall upto 50% is acceptable. The period required for 50% fall in sensitivity may be called as life span of the detector. Afterwards the detector should be replaced. If the sample quality is same and if the sample flow is around 100 ml/minute, the life span of the detector will be around 50 days.

The system was installed at RAPS-II. The life span of the detector was found only 15 to 20 days. Though the flow rate was adjusted at 100 ml/minute it was observed that under on-line conditions flow rate does not remain constant. Sometime the flow used to be very high and sometimes completely stops. Because of this the actual quantity of sample water passed through the detector could not be estimated. To check this erratic flow, it was decided to use a dedicated pump. It was also decided to pass flow to the detector with just 10 ml/minute while the sample flow through the sample conditioner kept at 100 ml/minute. This will improve the detector life span considerably.

At RAPS it was found that the measurement was affected by chemiluminescence due to chlorine added

for arresting the fungus formation in the sample pipe. When the charcoal column replacement was shortened to 15 days instead of a month, this problem disappeared. Another problem we encountered was due to interference of dissolved argon-41 in the sample water. The addition of bubbler had solved this problem completely. The bubbler is made of perspex tube of 1.2 cm diameter and 90 cm long. Its sample holding capacity is 100 ml. The bubbler is kept in such a way that even when there is no sample flow, water in the detector is not drained and air does not enter the detector. The bubbler also has a sample overflow line so that excess sample overflows to the drain line. The sample flow through the detector is also controlled to around 10 ml/minute.

To study the effectiveness of the bubbler, initially sample water was passed through the bubbler without bubbling the nitrogen gas. The reading was equivalent to 30 nCi/ml of tritium though the sample did not contain any tritium. Later, nitrogen gas was bubbled through the bubbler with a flow rate of 100 ml/minute. The reading has then fallen to 0.7 nCi/ml equivalent of tritium. When the nitrogen gas flow rate is increased to 200 ml/minute the reading has fallen almost to the background reading. For the flow rate of 400 ml/minute also the reading remained almost equal to background. Later, the study was repeated with compressed air. The bubbler was found to be effective in eliminating the dissolved argon even with compressed air. Since compressed air supply is readily available at site it was preferred.

With a new detector the minimum detection level at RAPS-II is found to be less than 1 nCi/ml. However, when the sensitivity falls to 50 %, the detection level will be still less than 2 nCi/ml. For a typical secondary water flow of 1400 litre/second and tritium activity concentration of 2 Ci/litre in heavy water, system will be able to detect the loss of heavy water of the quantity 5 litre/hour.

## 4. Conclusion:

The instrument installed at RAPS is working satisfactorily. It is being used continuously and is able to detect a heavy water leak less than 5 litre/hour. The efforts are being made to improve the sample conditioner so that the detector life span is increased. Efforts are also being made to improve the sensitively of the detector.

## Reference:

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