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AN ON-LINE TRITIUM-IN-WATER MONITOR

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The paper describes the development and operation of a continuous on-line tritium-in-water monitor for the detection of heavy water leaks into the secondary coolant light water of a heavy water power reactor. The heart of the instrument is its plastic scintillator sponge detector, made from 5 μm thick plastic scintillator films. The sponge weighs only about 1 g and is in the form of disc of 48 mm diameter and 8 mm thickness. The total surface area of the films is about 3000 cm^2. In the coincidence mode of counting, the detector gives 1000 cps for the passage of 3.7×10^4 Bq/cm^3 (1 μCi/cm^3) of tritiated water. The background in 6 cm thick lead shielding in the laboratory is 0.2 cps, and inside the reactor building it is below 1 cps. The monitor presently scans 18 sample lines in sequence for 5 min each and gives a printout for the activity in each line.

1. Introduction

Nuclear power reactors of heavy water type use large quantities of high purity heavy water as moderator and coolant. Operating conditions require the coolant heavy water lines to be maintained at high temperature and pressure. Light water is employed as the secondary coolant. Because of high temperature and pressure operation of the primary coolant lines, instances of line breakdown and consequent leakage of the heavy water into the secondary coolant light water are not uncommon. Instances of the moderator heavy water leaks have also been observed. Such moderator and coolant heavy water leaks have to be detected in the shortest possible time for initiating prompt corrective measures.

At the Rajasthan Atomic Power Station in India, detection of such leaks was carried out by measuring the tritium concentration in manually collected samples. Samples from various secondary coolant lines as well as the moderator heat exchanger were collected at 8 h intervals every day and the tritium concentration of the samples was assessed in the laboratory using a liquid scintillation counting system. The long gap of 8 h had to be kept because more frequent manual collection of several samples is impractical. This period, however, was considered too large because if any leak developed immediately after the sample collection, it could lead to loss of the precious heavy water. Only a continuous on-line monitor capable of detecting such leaks immediately could provide a satisfactory solution to this problem. With a commercially available instrument based on infrared absorption in which advantage is taken of differing infrared wavelengths for resonance absorption of H_2O, HDO and D_2O molecules, because of poor sensitivity, it was difficult to detect the heavy water loss at the site conditions unless the leak rate was high. For achieving higher sensitivity, it is desirable to detect the leaks with a tritium monitoring technique. Buildup of tritium in the heavy water takes place at a certain rate as a result of neutron activation of the deuterium atoms, which reaches a saturation value after several years.

In unit 2 of the Rajasthan Atomic Power Station, the present concentration of tritium in the coolant heavy water is about 1 Ci/kg. The light water secondary coolant flow rate in a line is typically 4000 l/min. A heavy water leak rate of 250 g/h (4 cm^3/min) shall produce a tritium concentration of 1 nCi/cm^3 in the secondary coolant of this line. With a sensitive on-line tritium-in-water monitor, it is possible to detect this activity. In a newly started heavy water reactor charged with fresh heavy water, a leak rate of about 1 kg/h could be detected after the reactor had remained in continuous operation for some months. As the tritium concentration builds up, the leak detection sensitivity improves proportionally. Because of the changes in the tritium concentration and possible variation in the secondary coolant flow rates, it may be difficult to correlate the monitor reading exactly with the heavy water leak rate on a long term basis. However, the important thing is to detect the leak itself. Exact assessment of the amount of heavy water leaked could be made subsequently. What is needed is a simple and relatively inexpensive tritium-in-water monitor that can be employed in on-line mode and can measure tritium concentrations of 1 nCi/cm^3, or lower, under site conditions.

In the early seventies, Osborne [1] in Canada re-

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ported the development of a flow cell detector packed with about 20 g of 0.0125 cm thick plastic scintillator sheets. Such sheets, though expensive, are available commercially. In the coincidence mode of counting, the detector gave 400 cps for the passage of 1 μCi/cm^3 of tritiated water. The background in the 10 cm thick lead shielding was 0.7 cps. The monitor described in this paper uses a simple and inexpensive flow cell detector packed with only 1 g of plastic scintillator material. Plastic scintillator obtained in the form of cheap ingots is easily converted into 5 μm thick flexible films of 125–150 mg weight. Seven to ten such film sheets are packed in a 50 mm diameter perspex flow cell in a particular way such that the packing gets converted into a spongy disc of 48 mm diameter and 8 mm thickness. Depending upon the number of film sheets used and the window width of the analyser, the detector gives a count rate of up to 1500 cps in coincidence mode for the passage of 1 μCi/cm^3 of tritiated water. The films could be converted into sponge form and used at will later. The life of the detector is affected only by the suspended matter in the sample water. With the use of suitable filters, it has been possible to prolong the life to several weeks. In 6 cm thick lead shielding, the detector shows a background of 0.2 cps in the laboratory. Inside the reactor also, the background is below 1 cps. Following the passage of high concentration tritium, no memory effect is noticeable after flowing 500 cm^3 of inactive water. The sponge permits water flow rates up to 500 cm^3/min.

2. Design of the detector and performance

The flow cell is made of perspex and consists of two parts, viz. the container and the lid. The two parts are separately encased in aluminium canning and each carries a nozzle. Leak proof assembly of the two parts is ensured by using a neoprene "O" ring and tightening by six symmetrically placed screws. Fig. 1 gives a sketch of the flow cell. The film sheets are packed in the container part.

The films are prepared on the surface of water. The technique to prepare thin plastic scintillator films has been described by Muga [2], by Batra and Shotter [3] and by Singh et al. [4]. As described by the latter, 3 g of plastic scintillator NE 102 is dissolved in a mixture of 90 cm^3 ethyl-acetate and 10 cm^3 amyl-acetate. 5 cm^3 of this solution containing 150 mg of plastic scintillator is poured in quick succession at the centre of a stainless steel tray of 30 cm × 30 cm × 4 cm (high), filled with about 3 l of distilled water. The film gets ready for removal in about 5 min. The film will be observed to be covered with a large number of randomly distributed holes of various sizes. The holes occupy about 40% of the total surface area. The film is carefully lifted from

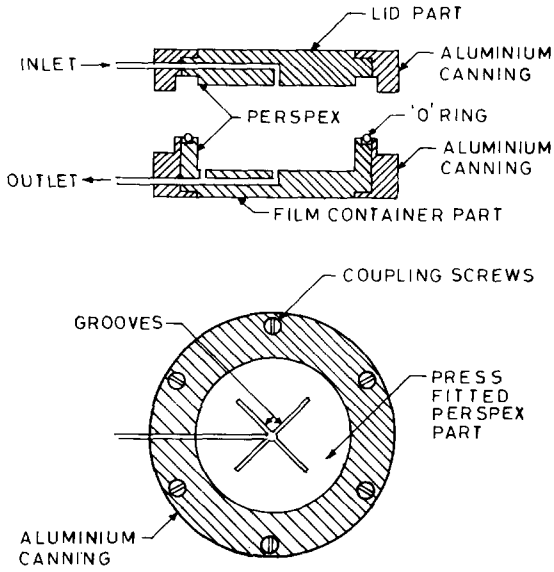

Fig. 1. Sketch of the flow cell detector.

the surface of water by stretching a clean sheet of white paper on the film and lifting the film along with the sheet. After removal the film is covered with another sheet of paper and allowed to dry. For filling in the detector, the dry film sheet is removed from the paper, folded and wetted in distilled water. It is then placed in the container part of the detector and is made to take the shape of the container with the help of a glass rod. Seven to ten sheets of the film are packed one over the other and the lid is tightened. The detector is now ready for use. If the detector is opened after some time, the filling will be found to have acquired the shape of a porous spongy disc. If necessary, this can be taken out and stored.

Out of 150 mg of plastic scintillator material in each film sheet, 20–30 mg of the film, mainly from the ends, gets lost during the preparation and the filling stages. The useful surface area, both faces inclusive, is about 500 cm^2 per film. The average thickness of the films is 5 μm.

The design of the detector is such that the water enters at the centre of the detector, spreads in various directions and flows through the sponge. The water inlet nozzle is in the lid part and the outlet nozzle is in the container part. Both nozzles face upwards and are placed close to each other. The holes linking the nozzles extend up to the centre of the lid and the container parts. With the help of suitable X-grooves in the lid and the container parts, the spread of water in various directions is assured. The resistance offered by the detector to the flow of water is low. In a seven sheet detector, a gravity pressure head of 1 m between the inlet and the outlet nozzles permits a water flow rate of about 100 cm^3/min.

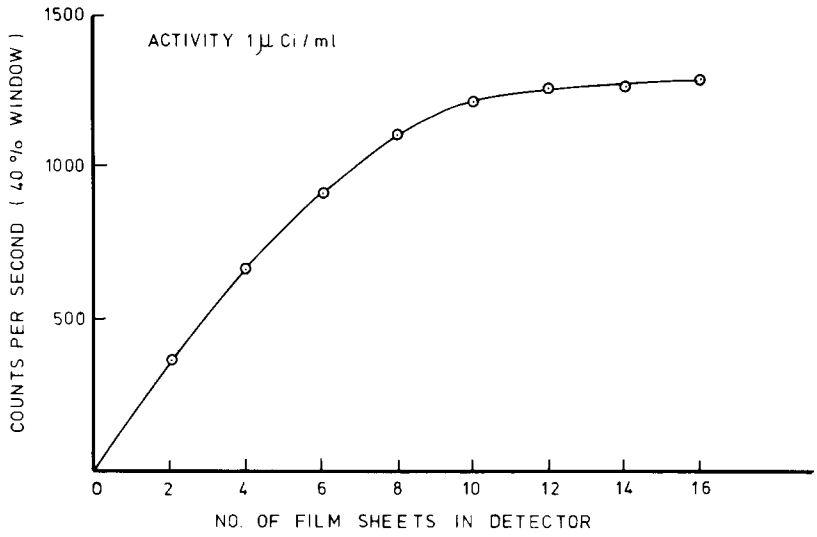

Fig. 2. Sensitivity as a function of the number of films.

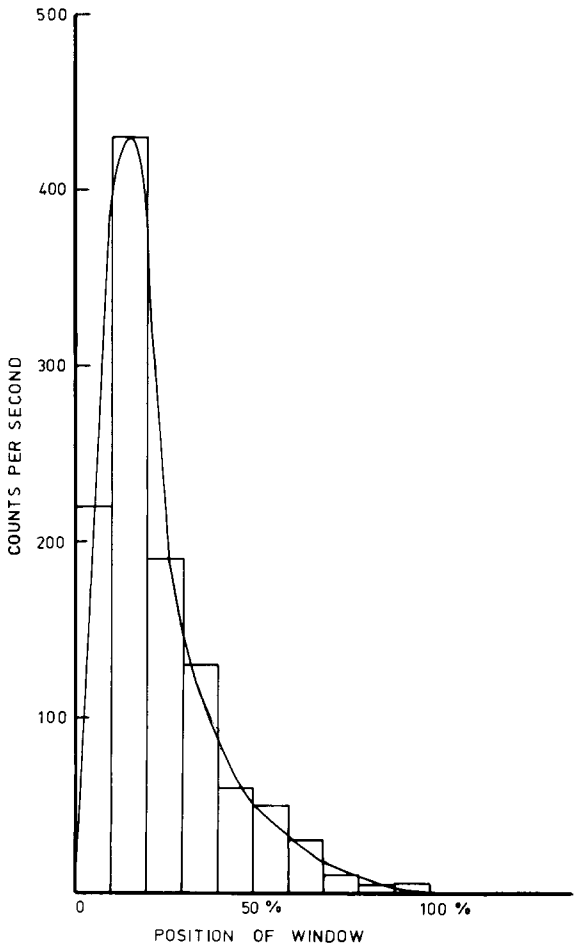

Fig. 3. Differential count rate at various window positions.

The ten sheet detector, under similar conditions, permits a flow of about 50 cm^3/min. Water flow rates up to 500 cm^3/min do not affect the quality and the shape of the packing.

The detector is coupled face to face to a matched pair of photomultipliers type EMI 9635 A, using silicone fluid as the optical coupling medium. The detec-

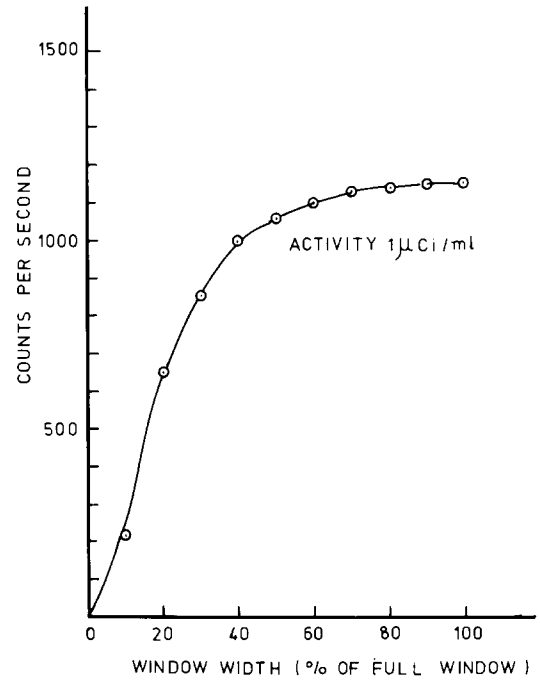

Fig. 4. Tritium sensitivity vs window width.

tor–photomultiplier assembly is enclosed in a light tight chamber measuring 49 cm × 10 cm × 11 cm (high) and is shielded with 6 cm thick lead. At optimum setting, the coincidence background is 0.2 cps in the laboratory. Fig. 2 shows the detector sensitivity as a function of the number of film sheets packed, for the passage of 1 $\mu Ci/cm^3$ tritiated water. The window width was kept at 4 V, i.e. 40%. Fig. 3 shows the differential count rate, i.e. the count rate contained in various 1 V window intervals between 0 and 10 V, such as 0–1, 1–2, 2–3 and so on. The detector used was packed with 7 sheets and the activity was 1 $\mu Ci/cm^3$. For the same detector fig. 4 shows the count rate as a function of the window width. At 40% window width, a 1 mR/h gamma field due to ^{60}Co gives 1.5 cps. The reproducibility of the results with a number of detectors is good. The sensitivity remains unchanged as long as the detector does not become dirty. The response is fast and at 100 cm^3/min sample flow rate, the full count rate is recorded within a minute. The flow of the sample heated up to 60°C did not show any appreciable change in the detector sensitivity.

3. The electronic assembly

The electronic circuitry consists of a high voltage supply for the two photomultipliers, amplifier and coincidence analyser, rate mater and local alarm. An associated electronic processor provides digital display, data printout and programmable command signals to the solenoid valves of the sequential sampler. The amplifier circuit has two independent amplifiers, one for each photomultiplier. Each amplifier output is simultaneously fed to a lower and an upper pulse amplitude discriminator. The low discriminator threshold is presettable and the upper one, which is helipot-controlled, can be varied between 0 and 10 V and sets the window width. After suitably shaping the lower and the upper discriminator outputs of the two photomultipliers, the anticoincidence output of each photomultiplier is obtained. The two anticoincidence outputs pertain to those pulses whose amplitude lies in the set window. In this way, large amplitude pulses of each photomultiplier that have a higher probability of non-tritium origin, are electronically eliminated. The two anticoincidence outputs are then fed to a coincidence circuit and the coincidence count rate is measured by the rate meter. The associated electronic processor receives the rate meter output and functions as programmed. A strip chart recorded display is also available.

4. Sample conditioning and the sequential sampler

The secondary coolant light water at the Rajasthan Atomic Power Station is drawn from a huge lake, fed by the river Chambal. The water looks very clear and, periodically, it is chlorinated to arrest the growth of algae. The detector, however, used to lose sensitivity very fast. A 125 mm glass fibre filter disc, sandwiched between Whatman filters 540 and 542, was used subsequently to pass filtered water through the detector. The filter pack, however, used to get choked in less than a day. The impurity was seen mainly on the glass fibre filter and looked greenish yellow. It was partly suspended matter of submicrometer range and partly organic matter. Whenever chlorination was carried out, the background tended to increase owing to chemiluminescence. Traces of ionic impurity also tended to increase the background. The filter assembly that was finally used consists of column of activated charcoal and mixed nuclear grade resin of 200 cm^3 volume, followed by a 30 cm diameter glass fibre filter sandwiched between Whatman filters 540 and 542. The filters are used in the pressure line and the sample flow rate through the filter is between 300 and 400 cm^3/min. The charcoal and mixed resin column remains effective for nearly a month. With this sample conditioner, the chemiluminescence and the ionic background have been eliminated. The life of the detector has also increased considerably.

Fig. 5 gives the block diagram of the heavy water leak detection system. The instrument is being used to monitor 18 lines sequentially, including two background lines. The lines are drawn from various locations through thin stainless steel tubings and the sample flow is under pressure. All the lines are terminated on a single panel named "D_2O to H_2O leak detection panel". Each line has a solenoid valve, operated by an electronic processor. The 18 lines have been divided into two groups of 9 lines each, the odd-numbered lines forming one group and the even-numbered lines forming another group. The first line of each group is the background line, carrying inactive water. Each group has its own sample tank of 400 cm^3 volume to which it supplies the sample water of its member lines sequentially for a period that can be preset between 1 min and 9 min. The two sample tanks, in rotation, feed the sample to the detector, each sample tank having its own solenoid valve, controlled by the electronic processor. The arrangement is such that at any time while one sample tank is feeding the detector the other sample tank is being filled with fresh sample from the next line. Presently the time is preset at 5 min. The role is reversed at the end of this period. During the period a tank is feeding the detector, the sample supply to this tank remains cut off. The sample feeding rate to the tank is between 300 and 400 cm^3/min and after the tanks get filled, the excess water overflows to the drain. This ensures the availability of the latest sample from a line. From the sample tank, about 300 cm^3 of the sample is passed through the detector at 75 cm^3/min and the balance is discharged into the drain.

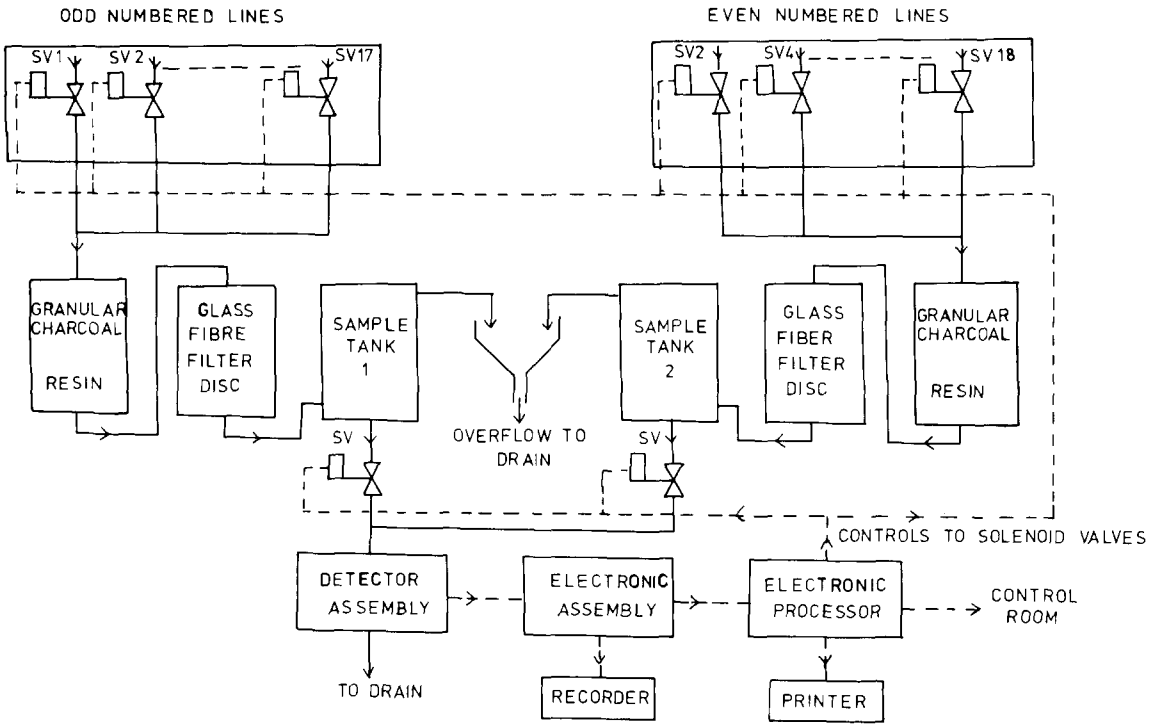

Fig. 5. Schematic of heavy water leak detection panel.

The flow of water from the sample tank to the detector is solely under gravity pressure.

5. Operational experience

The monitor was duly commissioned only in the first week of April 1984, after a continuous trial run for 5 months. No instances of heavy water leaks have been detected in this period. Liquid scintillation results have confirmed this. The monitor needs minimum attention. Filters are changed once a week and the charcoal and mixed resin column is changed once a month. About 5% reduction in the detector sensitivity due to gradual trapping of submicrometer particles has been observed in one month. It has been tentatively proposed to change the detector sponge once the sensitivity drops to 80% of the initial value. A spare probe assembly is available and this assembly having a fresh detector, could easily replace the initial one. The replacement of the sponge filling in the dirty detector can be carried out conveniently. A calibration check is carried out once in two week. For the present, the monitor alarm has been preset at 10 nCi/cm^3. No instances of false alarm have occurred so far. After watching the performance for some more time, the alarm point shall be progressively lowered.

A feature presently existing because of the use of only one monitor to scan 18 lines is that each line gets a turn after 90 min. By reducing the sampling time to 3 min, each line will get a turn in 54 min. However, since the dead volume of sample water in the lines and the filter is between 1 to 1.5 l and the sample flow rate through the 30 cm diameter glass fibre filter remains between 300 and 400 cm^3/min, 3 min sampling is not adequate. By using more than one monitor also the time could be reduced proportionally. This, however, will need substantial engineering changes in the sample supply system. As an additional measure, it is proposed to install one more monitor in the sump which collects water from all the lines before eventual discharge. The sample in the sump will be nearly 15 times more diluted as compared to the sample from any line where a leakage actually takes place. But any larger leak rate in any line could be detected within minutes even from the sump. With the installation of this monitor, it is hoped, detection of a D_2O leak in any line shall be possible long before substantial D_2O loss takes place. The authors are indebted to Dr. U.C. Mishra, Head, Air Monitoring Section, BARC for his keen interest in the development of the monitor. Thanks are due to Messrs M.G. Kadwani, A.G. Marker, M.C. Jain and M.V. Ramana Murthy for useful suggestions and help. Thanks are also due to Mr. K. Natarajan, Power Projects Engineering Division of the Department of Atomic Energy for his help in the calibration of the monitor and to Mr.

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