A SENSITIVE, REAL-TIME TRITIUM WASTE WATER MONITOR*

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HISTORY

Since its beginning over 30 years ago, Lawrence Livermore National Laboratory (LLNL) has maintained a constant environmental-surveillance program to ensure that concentrations of potentially hazardous materials in LLNL effluents are well below current guidelines. One of these effluents is the combined flow of sanitary and industrial waste water. Our waste-water-monitoring capabilities now include on-line low- and high-energy-photon detectors, a pH monitor, a x-ray fluorescence analyzer (XRFA) for metal ions, and a composite sampler. We are improving this system by developing a sensitive, real-time tritium waste water monitor. This monitor will quickly detect and mitigate either chronic or acute tritium spills to the sewer. Our early investigations have been reported previously.[1] We will summarize here the present system design and test results, and indicate directions for future investigations.

Our design goals for the monitor required the system to be--

- sensitive enough to detect 1-20 pCi HTO/ml (0.037 Bq-0.74 Bq. HTO/ml);
- capable of responding within four hours from the time tritiated waste water first enters the monitor;
- economical to operate on a continuous basis;
- rugged enough to operate at a remote environmental sampling station;
- easy to maintain on a routine basis;
- simple enough for a casual user to operate;
- capable of being connected to the existing microcomputer alarm system; and
- capable of obtaining a representative and particulate-free liquid sample.

SYSTEM DESIGN

Figure 1 shows the overall sample flow and electronic structure of the monitor system. It includes a macerator, ultraviolet (UV) sterilizer, "cross-flow" filter disk, passive mixing cell, flow cell detector, and dedicated microcomputer. The macerator reduces all foreign matter to 100 μm diameter or less in size. Because one of the largest maintenance problems is bio-fouling within the tritium monitor tubing, we installed a Model-500 ultraviolet flow sterilizer[2]; the sterilizer has allowed us to achieve the 16,000-μW-s/cm^2 dose recommended by the U. S. Public Health Service for sterilization of drinking water supplies for a few hours each day, and we have observed a reduction in the particulate hang-up within the "cross-flow" filter disk unit and associated tubing. The passive mixing cell is T-shaped; it surrounds the aqueous sample stream with a sheath of liquid scintillator, greatly reducing the chance of viscous gel formation. The dedicated LSI-11 microcomputer[3] makes the necessary quench correction and sends an alarm to a central emergency dispatch center if preset levels are reached. We divided our investigations of the system into two separate tasks: sample collection and radiation detection.

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SAMPLE-COLLECTION INVESTIGATION

We developed a prototype sampling filter of "cross-flow" design. The fast-moving stream in the instrument flume continuously washes across the filter medium, removing debris that would otherwise clog the filter. This cleaning action greatly extends filter life, while providing a clean sample. The prototype is installed in the existing XRFA flume; it consists of a 49-mm diameter filter disk, supported by a perforated stainless-steel backing, sealed with Neoprene O-ring gaskets to the back of the flume and held in place with a screw-on retainer ring (shown in Fig. 2).

We tried a variety of filter disks with pore sizes ranging from 10 μm down to 0.5 μm. (For reference, 1.2 μm is considered general filtration and 0.2 μm is considered sterilization.) The most important quality seems to be the macroscopic surface texture. Smooth-surface filters have lasted 10 times longer than rough-surface filters of comparable pore size. Also, smaller pore size filters lasted longer than similar large pore size filters. For example, 1.0 μm pore size filters lasted 70% longer on the average than identical 3.0 μm pore size filters. A 1.0 μm Teflon[4] filter disk, wetted with ethanol, has lasted the longest, approximately 10 days, at a filtrate flow rate of 0.23 ml/min. Visual inspection showed the filtrate to be particulate-free, thus fulfilling one of our most important design goals.

RADIATION-DETECTION INVESTIGATIONS

Liquid scintillation counting with a continuous flow cell appeared to be an optimum compromise between sensitivity and simplicity. The sample stream is mixed with a precise amount of liquid scintillation cocktail before being pumped through a flow cell detector. We set a design goal to limit cocktail consumption to one gallon (3.8 liters) per week to reduce operating costs.

There are a number of simple liquid scintillation, flow cell detectors available commercially. Most monitor high-specific-activity effluents from high-performance liquid chromatograph (HPLC) columns. We acquired a Flo-One, Model HP[5] radioactive flow detector on a long-term loan for proof-of-concept testing. The detector element is a thin-wall Teflon[4] tube coiled flat and sandwiched between two matched photomultiplier (PM) tubes. The usable cell volume is 2 ml. We characterized the performance of the unit as follows:

- The filtered waste water background count rate was 12% higher than the background count rate of drinking water. This was due to chemiluminescence and bioluminescence.
- The maximum counting efficiency for tritium in waste water was 10.6% as compared to 22.6% in drinking water. This was due to chemical quenching in the waste water.
- The minimum detectable activity (MDA) for a 20-minute counting period was calculated using the method of Tritium Measurement Techniques[6]. It was 265 pCi/ml (9.8 Bq/ml) with "known" (stable) background and 374 pCi/ml (13.9 Bq/ml) with "unknown" (variable) background.
- Response time for an aliquot of tritiated water containing 14 nCi (518 Bq) was less than 20 minutes to reach the MDA level, and 50 minutes to reach maximum count rate. Since at least four hours pass before LLNL effluent reaches the Livermore Water Reclamation Plant, sufficient time is available to evaluate the simulated emergency, and, if necessary, divert the flow to emergency holding basins at the plant.

FUTURE INVESTIGATIONS

We have begun to construct a second-generation monitor. It will include the following features to improve its sensitivity:

- A flat, spiral-wound coil of thin-walled Teflon[4] tubing sandwiched between the two matched PM tubes, similar to the flow cell developed by Schram and Lombaert.[7] The light-transmission properties of the cell will be improved by filling all voids with water or silicon oil, coupling the cell to the PM tubes with optical grease, and placing reflectors around the perimeter of the cell. Two PM tubes will be used in the coincidence counting mode. The flow cell and PM tubes will be placed in a graded metal shield to further reduce the background count rate.
- An external gamma source will be used for quench correction. The source will be actuated automatically by a simple mechanical system.
- Sachan and Soman have shown it is possible to decontaminate hydrophobic liquid scintillators.[8] We will decontaminate and reuse the collected "waste" scintillator, hence greatly reducing our operating costs.

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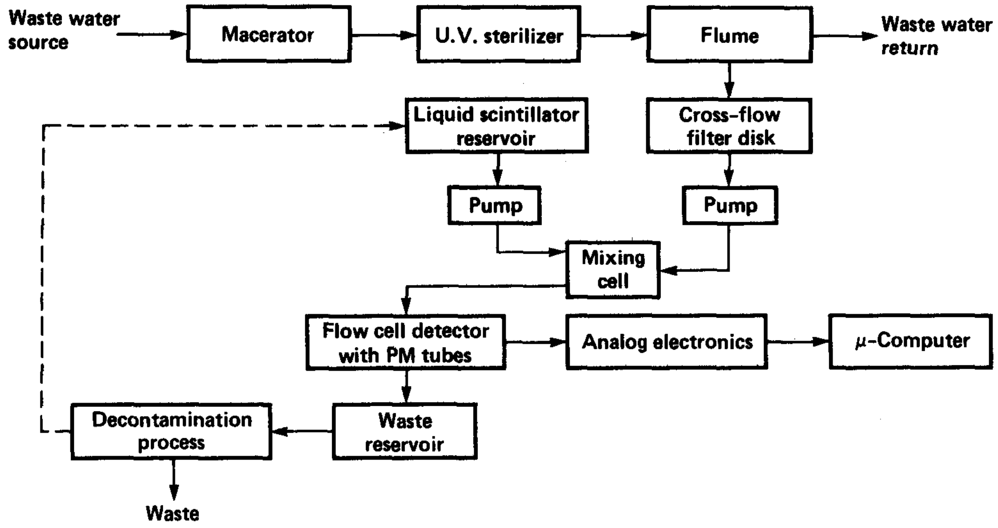

Figure 1. System block diagram.

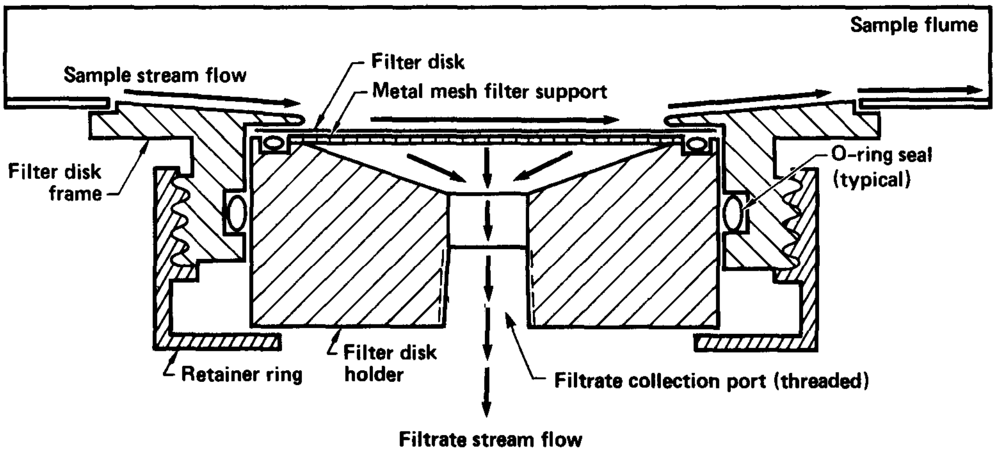

Figure 2. Cut-away view of the cross-flow filter.