WSRC-MS-96-0365
CONF-9606290--1

Development of a Remotely Operated, Field-Deployable Tritium Analysis System for Surface and Ground Water Measurement

by
K. J. Hofstetter
Westinghouse Savannah River Company
Savannah River Site
Aiken, South Carolina 29808
P. R. Cable

J. E. Noakes
University of Georgia
GA USA

J. D. Spaulding
University of Georgia
GA USA

M. P. Neary
University of Georgia
GA USA

M. S. Wasyl
Packard Instrument Company

A document prepared for 5TH ANNUAL LOW-LEVEL LIQUID SCINTILLATION COUNTING CONFERENCE at Yokohma from 06/20/96 - 06/21/96.

DOE Contract No. **DE-AC09-89SR18035**

DISCLAIMER

Development of a Remotely Operated, Field-Deployable Tritium Analysis System for Surface and Ground Water Measurement

John E. Noakes, James D. Spaulding, and Michael P. Neary
Center for Applied Isotope Studies, University of Georgia

Kenneth J. Hofstetter and Paula R. Cable
Westinghouse Savannah River Company

Michael S. Wasyl
Packard Instrument Company

Introduction

The environmental contamination resulting from decades of testing and manufacturing of nuclear materials for national defense purposes is a problem now being faced by the United States. Nationwide, an estimated 2.6 million curies of ~~radioactivity~~ mixed fission products has been released to the environment by a trio of facilities that reprocess nuclear fuel from production reactors - the U.S. Department of Energy (DOE) sites located in Hanford, Washington, Aiken (Savannah River), South Carolina, and Oak Ridge, Tennessee (Bradley et al., 1996).

At the Savannah River Site (SRS), the DOE facility responsible for the production of tritium and other special nuclear materials, there have been continuous low-level releases of tritium from prior disposal processes and unplanned events, which have affected ground and surface waters. As one of the most widespread and mobile contaminants found on site, tritium poses a potential environmental hazard to populations in nearby communities and in the downstream Savannah River watershed. The management of the high- and low-level radioactive wastes resulting from SRS activities is a problem of ongoing concern, particularly in the face of continued cutbacks in the funding available to support the environmental monitoring programs necessary for tracking and control of the waste streams. The characterization of tritium in ground and surface waters at the SRS has been ongoing for many years, conducted by manual sample collection and laboratory analytical methods. With present day advancements in computers, remote communication technologies, and low-background radiation detection systems, the components are now in place for the development of remotely operated, field-deployable analytical systems for near real time automated screening or monitoring of potential and existing contaminant plumes.

The Center for Applied Isotope Studies at the University of Georgia (CAIS), in cooperation with the Westinghouse Savannah River Company (WSRC) and Packard Instrument Company, have developed a prototype unit for remote, near real time, *in situ* analysis of tritium in surface and ground water samples. The Field-Deployable Tritium Analysis System (FDTAS) is sufficiently sensitive to measure tritium in water samples at

activities. The system is designed for remote control so that multi-site sampling, analysis, and data handling may be automated for unattended operations (Figure 1). By providing decision-quality data on a real-time basis from a mobile unit, the FDTAS will provide a cost-effective and more flexible alternative to the expensive and time-consuming methods of sample collection and laboratory analysis that are presently used for tritium monitoring at the SRS.

FDTAS Development

The goal of the FDTAS development program is to design and test a field-deployable system capable of rapid and remote liquid scintillation (LS) measurement of tritium in aqueous environmental samples. Specifications for the system included a background of no more than 3 counts per minute (cpm) and a counting efficiency of 25%, with a minimum detectable activity (MDA) of 10 Becquerels of radioactivity per liter (Bq/l) for a 100-minute count. Sample standardization, between-sample cleansing, and two-way remote communication capabilities were of primary importance for system development. The FDTAS is designed to allow a laboratory-based operator to instruct a remote unit (or units) to collect and analyze a sample, to reconfigure its mode of operation, to conduct an internal calibration, or to be interrogated for analytical results. Direct-dial modems provide the telephone communication and control link between the laboratory computer and a remote LS counter.

The Packard Radiomatic™ A 525TR LS counter (Figure 2) was selected as the basic instrument for low-level tritium measurement, but required a number of system modifications in order to meet the desired specifications. To accommodate measurement of the low levels of tritium expected in aqueous environmental samples, the aqueous sample volume had to be increased to 5 ml and measured on a flow/stop (batch) basis rather than on the continuous basis for which the 525TR was originally designed. The reduction of background was critical for the achievement of the lower detection levels specified, and was accomplished by the addition of lead shielding, the redesign of the counting chamber and sample vial holder, and the incorporation of $Bi_4Ge_3O_{12}$ (bismuth germanate, or BGO) windows in the counting chamber, coupled with after-pulse counting electronics..

The prototype FDTAS is now enclosed in a field-portable steel housing containing additional lead shielding (Figure 3). The redesigned counting chamber is made of high-purity electrolytic copper that is coated on the interior with a white reflective paint to enhance photon collection by the photomultiplier tubes. The chamber accommodates a specially designed 10 ml flow-through quartz sample vial of 8 mm thickness and 40 mm diameter. A pair of BGO windows, 6 mm thick, are incorporated in the counting chamber for reduction of background, and bracket both sides of the quartz sample vial. The counting chamber, quartz sample vial and BGO window configurations were optimized through a series of tests that were conducted using optical modeling software.

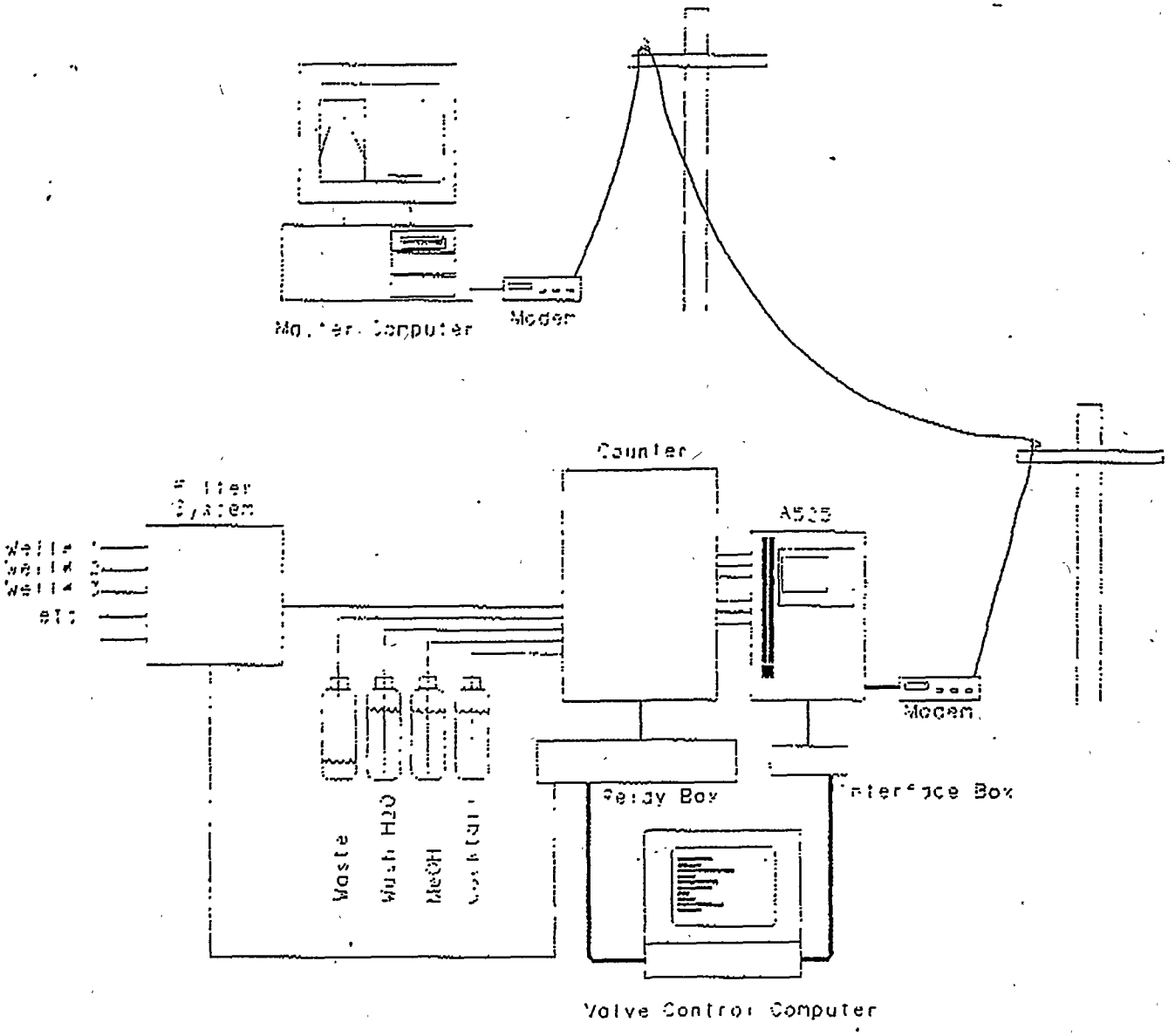
Modem
Counter
A535
Modem
Relay Box
Interface Box
Waste
Valve Control Computer

FIGURE 1.

To accommodate the need for multi-sample injection without sample cross-contamination, a flow/stop sample injection and system cleaning mechanism was assembled for the FDTAS using solenoid valves and metering pumps. This flow system assembly is controlled by a 24-channel relay accessory board and parallel digital I/O board in the valve control computer, permitting the collection of samples from multiple sites, with computer-automated sequencing of sample injection, cocktail mixing, sample counting, sample outflow, and cell cleaning cycles.

A range of LS cocktails, including Packard Ultima Gold LLT, Ultima Gold AB, Ultima Gold XR, and Ultima Flo M were evaluated for use with the FDTAS. In addition to the parameters of background and counting efficiency, the viscosity of the cocktail also required consideration. Sample backgrounds were determined using a Packard 2550 LS counter. The cocktails were then spiked with a known tritium activity and recounted to determine counting efficiencies. Ultima Gold LLT; normally a clear choice for tritium measurement due to a low background and high counting efficiency, was found to be too viscous to be readily washed from the sample vial during the cleaning cycle. The two least viscous cocktails, Ultima Gold XR and Ultima Flo M, were found to have significantly higher backgrounds and lower counting efficiencies when compared to Ultima Gold AB. Although Ultima Gold AB is a more viscous cocktail, it could nevertheless be washed fairly easily out of the sample vial, thus providing the best compromise for use with the FDTAS.

Using Ultima Gold AB, a series of mixtures having cocktail:water ratios of 40:60, 50:50, 60:40, and 70:30 were made up and counted to determine background. The mixtures were then spiked with a known tritium activity and recounted to determine counting efficiency. The 50:50 cocktail:water ratio was found to produce the best overall results for FDTAS tritium measurement.

Several tests were also conducted to maximize the efficiency of the flow-cell cleaning cycle to eliminate cross-contamination of samples. Of the alcohol solvents tested, methanol was found to produce the best cleaning results. Three washes of pure methanol, followed by three washes of dead (nontritiated) water serve to completely eliminate any sample "memory" in the system. The effectiveness of the cleaning cycle is demonstrated in Figure 4, which shows spectral data on sequentially measured samples including 1) the background on a nontritiated water sample; 2) a tritiated water standard (400 dpm); and 3) the background on a second nontritiated water sample following the methanol/water cleaning cycle.

FDTAS Testing and Evaluation

The remote sampling and analysis capabilities were tested at the CAIS laboratory in March of 1996. The master computer was set up in one laboratory and the LS counter and valve control computer were located in a separate laboratory. Using telephone/modem linkages, the sample collection/analysis/cleaning cycle was successfully operated from the remotely located master computer. A background of 1.35 cpm and a

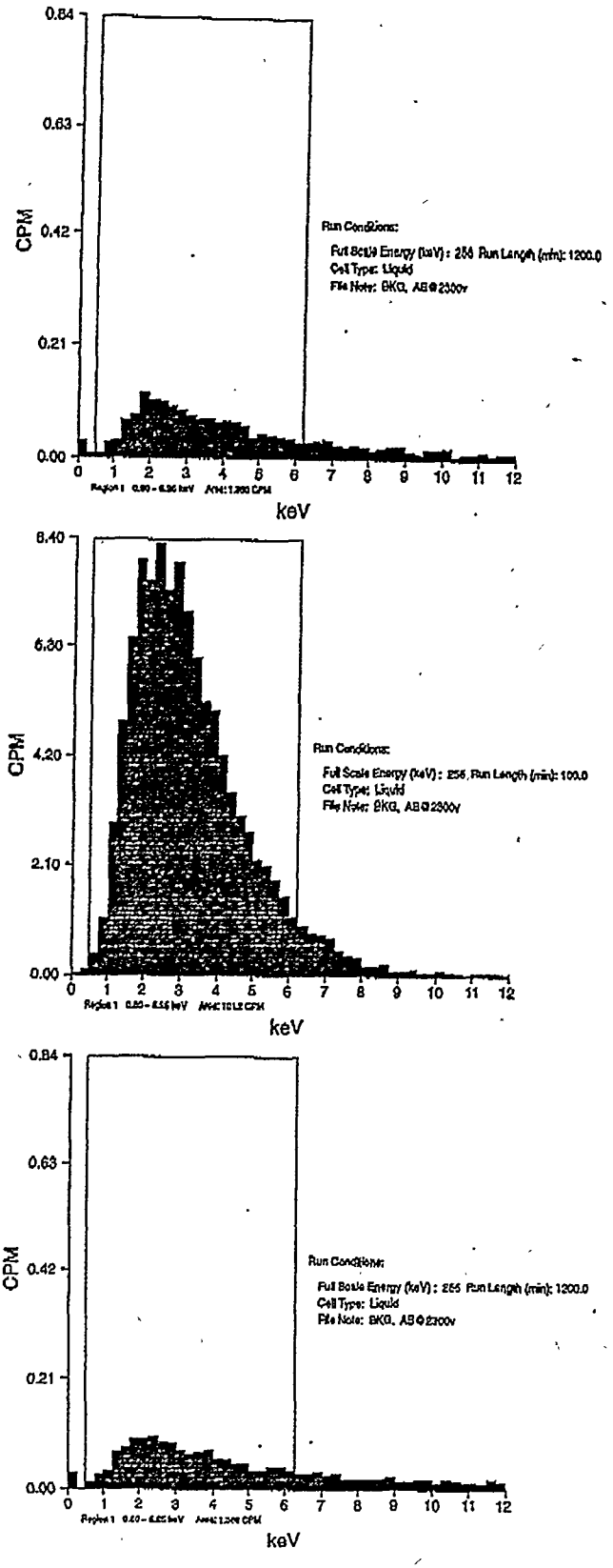
0.84
0.63
0.42
0.21
0.00
CPM
0 1 2 3 4 5 6 7 8 9 10 11 12
keV
Run Conditions:
Full Scale Energy (keV): 255 Run Length (min): 1200.0
Cell Type: Liquid
File Note: BKG, ABG 2300v
6.40
6.30
4.20
2.10
0.00
CPM
0 1 2 3 4 5 6 7 8 9 10 11 12
keV
Run Conditions:
Full Scale Energy (keV): 255 Run Length (min): 100.0
Cell Type: Liquid
File Note: BKG, ABG 2300v
0.84
0.63
0.42
0.21
0.00
CPM
0 1 2 3 4 5 6 7 8 9 10 11 12
keV
Run Conditions:
Full Scale Energy (keV): 255 Run Length (min): 1200.0
Cell Type: Liquid
File Note: BKG, ABG 2300v

FIGURE 4.

counting efficiency of 24% were demonstrated in this laboratory setting, yielding an MDA of less than 7 Bq/l for a 100-minute count and meeting the original specifications for the FDTAS.

A preliminary field test conducted on site at the SRS in May of 1996 was similarly successful. The FDTAS field unit was placed in a temperature- and humidity-controlled enclosure mounted on a small trailer for ready mobility (Figures 5 & 6). The test location selected for the FDTAS was "L" Lake, which was constructed in 1985 to receive the heated effluents of one of the five nuclear production reactors at the SRS. "L" Lake is located approximately 17 km from the central laboratory where CAIS and WSRC personnel set up the master computer. Remote communications between the laboratory and the field site were established using telephone and modem. Using the master computer, CAIS and WSRC personnel instructed the FDTAS to collect an aqueous sample, to acquire and analyze the sample spectrum, to evaluate background, and to empty and cleanse the sample vial in preparation for the next sample cycle. A background of 1.5 cpm and a counting efficiency of 24% were demonstrated in the field, which resulted in an MDA of 7.2 Bq/l for a 100-minute count.

For this preliminary test, the aqueous samples collected from "L" Lake were introduced directly into the FDTAS following a filtration and cleanup process performed by WSRC personnel using EI ChroM resins (Figure 7). WSRC is presently developing a fully-automated multi-port filtration system that will be integrated into the FDTAS, permitting remote sample acquisition from selected sample sites (*i.e.* specific wells within a cluster of wells) following commands issued through the master computer.

Ongoing Research

The two-year FDTAS development program is presently more than three-quarters complete. WSRC personnel are now directing efforts to the development and integration of a sample filtration system that will help reduce quenching agents in the aqueous samples before they are delivered to the FDTAS for analysis. The CAIS continues to work to develop a collimated ^{134}Ba or ^{241}Am external standardization capability for the FDTAS that uses a special sample vial and optically efficient geometric positioning. This capability is expected to result in more accurate tritium measurements through the correction of quench problems related to LS counting of tritiated water.

Extensive field testing of the completed FDTAS, including sample delivery and calibration capabilities, is scheduled to begin at the SRS in late July of 1996. Delivery of a second fully operational FDTAS instrument to WSRC is slated for September of 1996. The two systems will be used to conduct a joint and final test, for which the FDTAS units will be set up in different field locations and operated simultaneously from a central laboratory (Figure 8).

Conclusion

The Field-Deployable Tritium Analysis System has promise of wide application to areas of public concern such as the U.S. DOE nuclear fuel reprocessing facilities, which require a highly sensitive level of monitoring that cannot presently be achieved on a cost-effective basis. By permitting collection and analysis of aqueous samples on a selective, on-demand, and rapid basis, the costly routine sample gathering and lengthy laboratory analytical processes now used to monitor tritium in on-site wells and surface waters can be eliminated. The mobility and portability of the field unit, combined with the central laboratory operation of multiple units, enables monitoring programs to be conducted with far fewer personnel than is now possible.

With some modification of the system electronics, the FDTAS also has the potential for use as a combined alpha/beta/gamma counter. An alpha/beta/gamma counting capability, coupled with the remote field operational parameters of the FDTAS, would have a broad range of applications in environmental monitoring. Current LS counters can effectively differentiate and measure alpha and beta radiation through the use of pulse shape analyses. The pulses of the BGO detectors, presently used to discriminate against gamma events for background reduction, could conceivably be stored for a subsequent gamma analysis. This capability would permit rapid and remote measurement of anthropogenically released radionuclides such as ^{226}Ra, ^{228}Ra, ^{89}Sr, ^{90}Sr, ^{137}Cs, and ^{60}Co in addition to the measurement of tritium in surface and ground water samples.

References

Don J. Bradley, Clyde W. Frank and Yevgeny Mikerin, "Nuclear Contamination from Weapons Complexes in the Former Soviet Union and the ^United States", Physics Today, April 1996, pp 40-45.