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TRITIUM MONITORING OF GROUNDWATER AND SURFACES

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ABSTRACT

There are numerous facilities, both within the US and in the rest of the world, within the complex of radiation laboratories and production plants where tritium has been released into the environment because of historic or ongoing mission-related operations. Many of environmental restoration projects have detected low levels of tritium contamination in local streams, ponds, and/or ground water. Typically these waters are moving or have the potential to move offsite and are viewed as a potential risk to the public and environment. Los Alamos National Laboratory will modify the well-proven long-range alpha detection (LRAD) technique for detection of ionizing radiation to optimize a system for detecting tritium in groundwater and other surfaces. The LRAD technique relies on detection of ionized air molecules rather than direct detection of ionizing radiation. The detected electrical current is proportional to the number of ionized air molecules present, which is in turn a measure of the amount of contamination present. Although this technique has been used commercially to measure alpha contamination on objects and surfaces, the technique is also ideal for monitoring low-energy beta particles (such as those produced in tritium decay). We have demonstrated beta detection using ⁵⁴Mn, ¹⁴C, ¹⁴⁷Pm, ⁹⁹Tc, ⁹⁰Sr, and ³⁶Cl sources. Thus, the detector technology and detection of beta particles using this technology have both been demonstrated. The extreme short range of tritium beta particles (less than a millimeter in air) necessitates an optimization of the detector system. In this paper, we will discuss these new designs.

INTRODUCTION

The US Department of Energy (DOE) has several facilities within its complex of national laboratories and production plants where tritium has been released into the environment because of historic or ongoing mission-related operations. Many of the Environmental Restoration (ER) projects at these facilities have found low-level tritium contamination in local streams, ponds, and/or ground water. Typically these waters are moving or have the potential to move offsite and are viewed as a potential risk to the public and environment. The ER Integration Team, composed of members from 10 of the DOE's major facilities, identified near-real-time tritium monitoring of aquatic environments as the technology development need with the greatest immediate potential benefit. Across the DOE complex the environmental cleanup projects and stewardship (ES&H and WM) programs have had to address past (and in some cases ongoing) releases of radioactive and hazardous chemical constituents under a host of environmental regulations, and to increasingly stringent human health and ecological risk-based standards. At a number of facilities, tritium has been found in surface waters and/or groundwater at concentration levels that concern the regulatory authorities and the DOE. Once a tritium release has

entered surface or groundwater, it is usually not possible to eliminate the problem through a direct clean-up action. Consequently, management of the problem, including monitoring and tracking its migration/dispersion, becomes most important. Fortunately, tritium has a relatively short half-life (~12.5 yrs.) compared to many other radioactive elements and thus will decay to levels well below risk-based standards over several decades. During that period, it is important (and legally binding in many instances) that DOE have a means to effectively and efficiently monitor contaminated water on and under its facilities in order to ensure that it remain isolated from the public and other potentially impacted receptors. Currently, such monitoring is done by physically collecting water samples every few months, taking them to a laboratory for analysis, evaluating the results, and reporting the findings in an annual surveillance report, or some similar type of document. Sampling programs are expensive and will miss events that occur between (either in space or in time) the sample locations.

Los Alamos National Laboratory has the expertise and facilities to develop tritium sensors that will meet the DOE's long-term monitoring requirements for surface and ground water conditions, with near real-time measurements and remote operations capability. (1) Our goals include:

- 1. establishing the technical feasibility of quantifying tritium concentrations in water by measuring beta radiation in an air space immediately above the water surface;
- 2. developing and testing specific designs optimized for stagnant water, flowing water, and in well bores; and,
- 3. linking these detectors to remote transmission equipment.

TECHNIQUE

The LRAD technique relies on detection of ionized air molecules rather than direct detection of ionizing radiation. (2, 3) As illustrated in Fig. 1, all ionizing radiation produces ions as the incident radiation loses its energy in ambient air. Ions generated in the air above a contaminated surface (such as water) can be attracted to an ion collector and the current associated with this ion movement registered as a small current. This current is directly proportional to the number of ions produced; the number of ions is directly proportional to the strength of the radioactive contamination. Thus, the detected current is a measure of the amount of contamination present.



Fig. 1. Ambient air ionization method for detecting radioactive contamination. Ionizing radiation (a, b, or γ) from the contaminated surface ionizes molecules of ambient air. These ions are collected and measured as an electric current.

The LRAD technique, as illustrated in Fig. 1, can easily be applied to measuring contamination on a surface. In this type of LRAD detector (termed an electrostatic detector) the air ions are attracted to the collection electrode by an electric field. A simple electrostatic LRAD monitor is shown in Fig. 2. All ions generated in the detection chamber are attracted to the Ion Collector electrode by a voltage applied to this electrode. The current from this electrode to ground is proportional to the number of ions in the chamber and hence to the amount of contamination present on the surface.



Potentially Contaminated Liquid or Surface

Fig. 2. A simple surface monitor based on the concept illustrated in Fig. 1. All ions created within the detection chamber are attracted by an electric field to the ion collector. This movement of ions creates a small current $(\sim 10^{-14} \text{ A})$ that can be detected externally.

DETECTOR IMPLEMENTATIONS

This technique has been used extensively to measure alpha contamination on objects and surfaces. The LRAD technique is ideal for monitoring alpha contamination due to the short range (several centimeters) and high density of ionization of the alpha particle. Low-energy beta particles (such as those produced in tritium decay) also have a short enough range to be detected with LRAD-type monitors. We have demonstrated beta detection using ⁵⁴Mn, ¹⁴C, ¹⁴⁷Pm, ⁹⁹Tc, ⁹⁰Sr, and ³⁶Cl sources. Thus, the detector technology and detection of beta particles using this technology are both demonstrated facts. The extreme short range of tritium beta particles (less than a millimeter in air) necessitates an optimization of the detector system. In this paper, we will discuss these new designs as well as existing results.

An example of a tritium-optimized surface monitor is shown in Fig. 3. The signal collection electrode in this detector operates in the same fashion as the simple surface monitor shown in Fig. 2. However, a second detection region and collector plate have been added immediately behind the first. This 'background' collection volume is **not** exposed to the contaminated surface, but **is** exposed to the same air as the front volume. (4) Thus, the background volume will be sensitive to gamma backgrounds and airborne radon. These backgrounds can be removed from the signal by subtracting the background result from the signal result. Following background subtraction, this detector should only be sensitive to the surface contamination.



Fig. 3. Tritium-optimized surface monitor. Two methods are employed to reduce the radon background. 1) The detection chamber is made very thin so that most of an alpha particle's energy is lost in the collector plate while all of the tritium-beta's energy is deposited in the air. 2) The current from the background chamber (caused by airborne radon) is subtracted from the current from the signal chamber.

A related implementation of the surface monitor is shown in Fig. 4. In this case the collection electrode has been wrapped into a cylinder. This cylinder is dropped down the center of a borehole. Again, as in previous examples, this detector measures all contamination on the surface – in this case, the inner surface of the borehole.



Fig. 4. Implementation of the surface detector geometry illustrated in Fig. 2 in a configuration that is suitable for *in-situ* borehole measurements.

BACKGROUND SUPPESSION

The sensitivity of LRAD-based detectors is sufficient to detect tritium at the required low levels. (5) However, LRAD-based detectors are intrinsically very good alpha particle detectors as well as beta detectors and a typical alpha particle has 1000 times as much energy as a typical tritium beta. Thus, consideration must be given to the suppression of ambient alpha signals as well as to sensitivity to beta signals. In particular, since these detectors use ambient air as a detection medium, radon decays in this air are a significant issue. We have developed three ways to suppress the background alpha response relative to the tritium beta response. These are presented in order of increasing effectiveness.

Alpha particles have a range of several centimeters in air while the tritium beta will only travel a millimeter or less. Thus, an alpha particle will loose a large part of its energy in the collector plate of a very thin detector. However, the tritium beta will deposit all of its energy into the air of the detector. In addition, the thin detector will enclose a smaller volume of ambient air, reducing the amount of radon that is present in the detector.

Neither of the effects will eliminate the signals due to radon by themselves. However, these will reduce the contributions due to radon gas and can be used in conjunction with the other methods to eliminate the spurious radon signals.

A second detection chamber can be built immediately behind the first. This second, or background canceling, chamber will detect backgrounds caused by gamma fluxes or atmospheric radon but not the intended signal due to surface contamination. As described in Ref. 4, the signal from the second chamber is subtracted from the first to generate a surface contamination measurement that does not include background signals. This technique is powerful enough that it has been included explicitly in the surface monitor illustrated in Fig. 3.

The pulse counting detector described in Ref. 5 has proved to be very effective in separating alpha and beta signal when the alpha counting rate is low. Individual alpha particles generate very large pulses in the detector that can be easily distinguished from the more uniform beta-generated background level. Although this technique has been used to monitor alpha signals in the presence of a fluctuating background, it could equally well be used to measure a fluctuating 'background' signal caused by tritium in the presence of alpha decays. In the first instance the pulses are kept and the background is discarded. In the second instance, the background will be kept and the pulses discarded.

CONCLUSIONS

Both the detector technology and detection of beta particles using this technology have been demonstrated. Systems intended for beta measurements will have to incorporate one or more background cancellation methods. In addition, the extreme short range of tritium beta particles (less than a millimeter in air) will necessitate an optimization of the detector system that is different from the existing alpha-optimized systems.

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