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Tritium on Surfaces*

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ABSTRACT

We describe development of small passive, solid-state detectors for in-situ measurements of tritium, or other weak beta-emitting radionuclides, on surfaces. One form of detector operates on the principle of thermally stimulated exoelectron emission (TSEE), the other by discharge of an electret ion chamber (EIC). There are currently two specific types of commercially available detector systems that lend themselves to making surface measurements. One is the thin-film BeO on a graphite disc, and the other is the Teflon EIC. Two other types of TSEE dosimeters (ceramic BeO and carbon doped alumina) are described but lack either a suitable commercially available reader or standardized methods of fabrication. The small size of these detectors allows deployment in locations difficult to access with conventional windowless gas-flow proportional counters. Preliminary testing shows that quantitative measurements are realized with exposure times of 1-10 hours for the TSEE dosimeters (at the DOE release guideline of 5000 dpm/100cm² for fixed beta contamination). The EIC detectors exhibit an MDA of 26,000 dpm/100cm² for a 24 hour exposure. Both types of integrating device are inexpensive and reusable. Measurements can, therefore, be made that are faster, cheaper, safer, and better than those possible with baseline monitoring technology.

INTRODUCTION

A comprehensive review article on tritium monitoring techniques was published in the December 1993 issue of the Health Physics Journal¹. Somewhat surprising was the lack of mention of passive tritium monitoring techniques that have had a variable history of attempted development over the past two decades. An evaluation of past and current work with TSEE dosimeters and EIC's was necessary for rejuvenating our own research on tritium surface monitoring. We are working closely with other researchers and taking advantage of their more recent advances.

Pioneering work with TSEE for dosimetry began at ORNL in the 1970's under the direction of Becker and Gammage. The aim was to produce personnel dosimeters that could measure and discriminate between penetrating and non-penetrating ionizing radiations. The state-of-the-art material at that time was a ceramic BeO known by its trade name Thermalox 995. Gammage and Cheka successfully monitored tritium with Thermalox 995². For long-term personnel monitoring Thermalox 995 experienced stability problems due to the condensation of water vapor if the ambient temperature fell below the dew point. The most notable setbacks were a ruined TSEE peak shape and a loss of TSEE intensity³.

In the 1980's, German researchers developed a thin-film BeO on a graphite substrate that reportedly overcame the stability problems linked to hydration. Kriegseis et al. used thin-film BeO to measure tritium beta doses spanning five orders of magnitude⁴.

A new reader for TSEE analysis was commercialized in the late 1980's by Fimel (France). It is the result of 12 years of counter development at the Commissariat à l'Energie Atomique, France. The Fimel detection scheme uses a multipoint Geiger counter with cathodic focusing to reduce the normal dead time of a Geiger counter from 25 μ s to 2 μ s⁵. We are currently using this TSEE reader.

In the early 1990's, a third type of TSEE dosimeter was developed by the Russians. It is carbon doped alumina (α -Al₂O₃:C). The α -Al₂O₃:C is reported to be 20 times more sensitive to X and gamma radiations than thin-film BeO⁶. A response to tritium beta rays was not measured.

The EIC studied in this work is a modification of the E-PERM® (electret passive environmental radon monitor)⁷ commercialized by Rad Elec Inc. of Frederick, Maryland, USA. The EIC was originally

developed for radon monitoring with subsequent adaptations permitting measurements of tritium vapor and surface alpha contamination⁸. Here we have further adapted the E-PERM® for making measurements of tritium surface contamination. Development work was established through a cooperative research and development agreement (CRADA) between Rad Elec Inc. and ORNL.

EXPERIMENTAL

Materials

TSEE dosimeters. BeO thin-film dosimeters (0.38cm^2) are produced by the Staatliches Materialprüfungsamt Nordrhein-Westfalen (Germany) on special order. Dosimeters are prepared by evaporation of a 150 nm film of beryllium onto a graphite substrate followed by oxidation in wet nitrogen at 1300°C ⁹. The ceramic BeO Thermalox 995 (1.22cm^2) is commercially available from the Brush Beryllium Co., Elmore, Ohio, USA. Thermalox 995 dosimeters were preconditioned according to the stipulations of Gammage and Cheka³. Prototype $\alpha\text{-Al}_2\text{O}_3\text{:C}$ dosimeters (1.22cm^2) were fabricated by Durham and Akselrod⁶ by cold pressing carbon-doped aluminum oxide powders of varying grain sizes onto aluminum substrates.

EIC. The EIC detectors were provided by Rad Elec Inc., Frederick, Maryland, USA. For surface tritium monitoring, the EIC configuration is a 3.8mL ionization chamber containing a 9.5cm^2 charged Teflon electret. The effective source-to-detector distance is 4mm, which approximately matches the average range of tritium beta rays in air.

Tritium source. The NIST traceable calibrated solid tritium source used in these experiments is a 0.3mm thick layer of anodized aluminum foil. The beta emission rate is 81 β/sec from an active area of 4.9cm^2 .

Holders and Exposures

TSEE dosimeters. The thin-film BeO lies within a 1mm indentation on the graphite disc (Fig. 1). The indentation serves to protect the thin-film from abrasion or directly touching any tritiated surface that is being monitored. If the surface being measured contains removable tritium, a thin disposable washer should be used to prevent the transfer of tritium contamination to the graphite disc. The washer was also used when experimenting with Thermalox 995 and $\alpha\text{-Al}_2\text{O}_3\text{:C}$ dosimeters. The washer is analogous to a device used by Gammage and Cheka³ (Fig. 2).

EIC. The EIC is made of a holder and a spacer which screw together. During exposure the spacer and holder protect the Teflon electret from being touched or contaminated. Exposures are made by inverting the TSEE dosimeter or the EIC so that the sensitive region faces the surface in question. Exposures were conducted for times of several minutes to several days.

Reader and Recording

TSEE dosimeters. Incident tritium beta rays cause ionization in the surface region with subsequent population of stable near-surface trapping centers. Upon thermal stimulation, liberated electrons escape the surface with a few keV of kinetic energy. The exoelectrons are counted as a function of substrate temperature with the Fimel reader and displayed as a characteristic "glow" curve (Fig. 3). The area under the principal TSEE peak is proportional to the tritium beta exposure.

EIC. The static positive surface charge of the EIC (Fig. 4) attracts the negative ions generated in air by absorbed beta rays. The collected ions reduce the Teflon surface charge which is read as a voltage drop with a portable electrometer. The rate of voltage drop is proportional to the surface tritium activity. To make readings, the Teflon in its holder is unscrewed from the spacer and inserted into the portable voltage reader.

RESULTS AND DISCUSSION

TSEE dosimeters

BeO thin-film. The thin-film BeO dosimeters on electrically conductive graphite substrates were found to be the most reliable. These dosimeters responded linearly to tritium activity over nearly three decades of integrated exposure (Fig. 5). The number of recorded exoelectrons per incident beta ray was

0.27±0.07 which is more than twice the response obtained by Kriegseis and colleagues⁴ using a monopoint reader.

Thermalox 995. An early saturation in response, probably due to positive charging of the emitting surface, was observed with Thermalox 995 (Fig. 6). In contrast, Gammage and Cheka³ observed a near linear response to a tritium contaminated aluminum ring (Fig. 7) over several decades of integrated exposure using a different reader. This reader electrically grounded the emitting surface thus minimizing charge buildup and reducing suppression of exoelectron emission. Our Fimel TSEE reader does not have that grounding capability which may explain the premature saturation of response. Using data from nonsaturated responses, the number of recorded exoelectrons per incident beta ray was 0.26±0.07 which indicates that the intrinsic response of Thermalox 995 is reproducible and nearly identical to that of thin-film BeO.

α -Al₂O₃:C. The α -Al₂O₃:C dosimeters produced a linear response over nearly three decades of integrated exposure to surface tritium. These experimental detectors have a response that is very dependent on grain size. Dosimeters having larger grains yielded a higher efficiency at the expense of decreased reproducibility. The average number of recorded exoelectrons per incident beta ray for many samples was 0.08±0.05. The higher sensitivity of α -Al₂O₃:C to penetrating radiation does not extend to weakly penetrating beta rays. Therefore, at this stage in development, it is questionable if α -Al₂O₃:C will produce a more sensitive dosimeter than thin-film BeO. Certainly more developmental work will be needed to produce optimized dosimeters with reproducible characteristics.

Table 1 gives necessary exposure times for TSEE dosimeters to quantify tritium surface contamination at the current DOE beta surface contamination release limit of 5000 dpm/100cm² at a 3:1 signal-to-noise ratio. Although thin-film BeO and Thermalox 995 dosimeters have nearly identical efficiencies, the area of the Thermalox 995 detectors is 3.2 times that of the thin-film BeO detectors. Therefore, in order to quantify the same surface activity, thin-film BeO requires exposure times that are 3.2 times longer than Thermalox 995.

EIC

The EIC response is linear for exposure times of minutes to tens of hours (Fig. 8). EIC's were exposed to a large-area tritium source (representing an infinite planar source) with a surface emission rate of 16.9 β /cm²-sec. Under field conditions, a practical lower limit for quantifiable response is a 10V drop after an exposure of 24 hours. The corresponding surface tritium activity to produce this electret voltage drop is 26,000 dpm/100cm².

Table 1. Comparison of exoelectron detectors

Detector	Exposure Time ^a (hours)	Intrinsic Efficiency (exoelectrons/beta)
BeO thin-film	2.2	0.27 ± 0.07
Thermalox 995	0.7	0.26 ± 0.07
α -Al ₂ O ₃ :C	4-10	0.08 ± 0.05

^a Time necessary to quantify 5000 dpm/100cm²

CONCLUSIONS

BeO thin-film TSEE dosimeters and the EIC detectors are well suited for monitoring total tritium (fixed and removable) on surfaces. Both are small, enabling them to be deployed in locations difficult to access with windowless gas-flow proportional counters. However, thin-film BeO dosimeters are not routinely produced and can only be obtained on special order. The hard, rugged ceramic BeO Thermalox 995 also performs adequately but will need a modified reader to prevent severe nonlinearity of response due to surface charging. The prototype α -Al₂O₃:C dosimeters need further development work before they can be adequately evaluated. The EIC commercialized by Rad Elec Inc. is easy to use and is less sensitive to tritium beta rays than TSEE dosimeters. The apparent applications for both TSEE dosimeters and the EIC detectors will be in decontamination and decommissioning of tritium processing facilities which cannot be monitored for total tritium with current baseline technology. Also, as fusion reactor technology matures, handling and storage of large inventories of tritium will lead to inevitable contamination problems. Field tests are scheduled in the near future at ORNL and Savannah River.

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Figure 1 BeO thin-film dosimeter.

Figure 2 Thermalox 995 disc and holder (reference 2).

Figure 3 Thermally stimulated exoelectron emission glow curve.

Figure 4 Electret Ionization Chamber.

Figure 5 BeO thin-film linear response to tritium source (beta emission rate = 6/sec).

Figure 6 Saturation of Thermalox 995 response due to charge buildup using Fimel reader (beta emission rate = 20/sec).

Figure 7 Response of Thermalox 995 with grounded reader of Gammage and Cheka (reference 2).

Figure 8 EIC tritium response curve (beta emission rate = 81/sec).