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Monitoring of Tritium-Contaminated Surfaces, Including Skin

Mesure sur surfaces contaminées par le tritium, dont la peau

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RÉSUMÉ

Nous avons examiné divers moniteurs de contamination de surfaces par le tritium, qu'on trouve dans le commerce, ainsi que différents matériaux proportionnels et techniques de mesure directe et indirecte par essuyage de surfaces contaminées, dont la peau. Aucune des méthodes ou aucun des instruments évalués n'a été plus sensible que la méthode de comptage au scintillateur liquide (LSC). Les méthodes de mesure aux compteurs proportionnels à fenêtre ouverte ont été en général plus sensibles que celles aux LSC de moins d'une demie fois mais offrent les avantages de fournir les résultats presque tout de suite et de ne nécessiter aucune préparation d'échantillons.

Le PC-55 de la Nuclear Measurement Corporation est l'instrument le plus commode que nous avons éprouvé pour l'analyse de la mesure courante par essuyage. Le PC-55 a été plus sensible que le LSC d'environ un tiers lorsqu'on l'a utilisé avec les matériaux à papier filtrant d'Ontario hydro. On peut obtenir des résultats de mesure de contamination de surfaces en quelques minutes avec le PC-55 alors qu'il faut quelques heures avec le LSC. La sélection de matériaux d'essuyage pour utilisation avec des compteurs est critique. Un matériau d'isolation électrique peut produire une charge électrostatique sur sa surface; ceci peut modifier le gradient de champ du détecteur et influencer défavorablement les résultats. Bien que le PC-55 soit assez sensible et très commode, il faut avoir de l'expérience en utilisation de celui-ci avant de la recommander à la place des méthodes de comptage au scintillateur liquide. La sensibilité du PC-55 à la contamination interne par le tritium peut limiter son utilité.

Du fait de la complexité de l'utilisation d'animaux vivants pour évaluer les méthodes de mesure directe et indirecte de la contamination de la peau, on a étudié la peau de porc comme remplacement possible. Nous en avons conclu que, pendant les quelques premières heures après l'exposition, la peau de porc permet de simuler le processus de transfert des contaminants à la peau de l'animal qui est entrée en contact avec une surface contaminée par le tritium.

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ABSTRACT

We have examined various commercially available tritium surface contamination monitors along with different swipe media and techniques for direct and indirect (swipe) monitoring of contaminated surfaces, including skin. None of the methods or instruments evaluated were more sensitive than the swipe and liquid scintillation counting (LSC) method. Swipe measurements with open-window proportional counters were, in general, less than half as sensitive as LSC, but have the advantages of having the results available almost immediately, and no sample preparation is required.

The Nuclear Measurement Corporation's PC-55 is the most suitable instrument we tested for the analysis of routine swipe measurement. The PC-55 was about one third as sensitive as LSC when used with Ontario Hydro filter paper swipe media. Surface contamination measurement results can be obtained within minutes using the PC-55, compared to hours using LSC. The selection of swipe media for use with proportional counter-based instruments is critical. A medium that is electrically insulating can develop an electrostatic charge on its surface; this may alter the field gradient in the detector and may adversely influence the results. Although the PC-55 is sufficiently sensitive and very convenient, operational experience with the instrument is needed before recommending that it replace current LSC methods. The PC-55's susceptibility to internal tritium contamination may limit its practical usefulness.

Because of the complexity of using live animals to evaluate direct and indirect methods for assessing skin contamination, pig skin was investigated as a possible substitute. We concluded that, for the first few hours post-exposure, pig skin mimics the kinetics of animal skin that has contacted a tritium-contaminated surface.

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EXECUTIVE SUMMARY

For operational radiation protection, removable tritium surface contamination is the quantity of interest, because it represents the amount of tritium that could be transferred to workers, under normal working conditions. It is assumed that this component of tritium surface contamination is identical to that which can be measured by a routine smear survey. Typically, a 100 cm^2 area is swiped with a suitable media, and the activity removed by the swipe is counted using a liquid scintillation counter.

Although the swipe and liquid scintillation counting (swipe/LSC) method is very sensitive, the results of which are easy to interpret, it requires sample preparation and the results are usually not available immediately. In addition, the liquid scintillation counter is normally located in a part of the facility other than the workplace to be monitored. An instrument or technique that is as sensitive as swipe/LSC, but which requires less preparation time and provides results more quickly, would be beneficial.

We have examined several commercially available tritium surface contamination instruments and techniques. No instrument or technique is more sensitive than swipe/LSC. The PC-55, 2π proportional counter, from Nuclear Measurement Corporation, is about one third as sensitive as the LSC method for analyzing tritium-contaminated swipes when used with Ontario Hydro filter paper swipe media. The selection of swipe media for use with a proportional counter-based instrument is critical. Media that are electrically insulating can develop an electrostatic charge on the surface of the swipe, which may alter the electrostatic field gradient in the detector and thus the counting efficiency. The swipe media should be somewhat conductive and not so absorbent as to decrease the beta emission rate by self-absorption.

The limited dynamic range, and susceptibility to contamination, of the Berthold LB1210 surface-contamination monitor with an LB6255 windowless proportional counter restricts its usefulness to relatively clean laboratory environments. The Whitlock VSC 5000 requires a large, smooth, flat, non-porous, and non-transparent surface for proper operation. Although very sensitive and portable, this instrument has limited usefulness for routine workplace surface monitoring.

The Herfurth Microcont and the Berthold LB1210 with a BZ-200 XK-P probe are not sufficiently sensitive for routine workplace surface monitoring. A prototype surface contamination monitor based on electrets requires too long a sampling time for low-level tritium contamination monitoring. MeltiLex™ , a wax-based plastic scintillant, may offer a convenient alternative to LSC counting. Measurements with a prototype monitor, with two photomultipliers in coincidence, using MeltiLex™ discs gave results comparable to swipe/LSC. The technology is currently available to make a commercial swipe monitor using MeltiLex™ scintillant. The availability, durability and cost of the MeltiLex™ sheets would have to be investigated further before a monitor based on this medium could be recommended for routine use.

™ MeltiLex is a registered trademark of Wallac Oy., Turku, Finland.

Although our main focus was on measuring tritium-contaminated stainless-steel surfaces, some tests were done on skin. It is possible to detect activity on skin that has come in contact with tritium-contaminated surfaces. Results with the Berthold LB1210 were difficult to interpret, due to the limitations of the probes used, and uncertainties in the surface contamination.

Pig skin was investigated as a possible substitute for live animals, with respect to tritium skin contamination. Preliminary results indicate that pig skin may simulate, for the first few hours post-exposure, the kinetics of animal skin that has contacted tritium-contaminated surfaces. The use of a standard surface for tritium contamination and to conveniently simulate skin would be advantageous for instrument evaluation.

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1. INTRODUCTION

Studies by Hutchin and Vaughan (1963), Eakins et al., (1975), Johnson et al., (1988), and Trivedi (1989, 1992 and 1993) have shown that if skin comes into contact with surfaces that have been exposed to high concentrations of HT gas, significant uptake and retention of tritium occurs in the skin, as well as in other organs. An accurate picture of the amount and form of tritium sorbed onto and into contaminated surfaces, and how it interacts with other systems, is important for contamination and dosimetric calculations.

A significant amount of work has been done on the nature of the surface-bound tritium on stainless steel (Hirayabashi et al. 1984a; Hirayabashi and Saeki 1984; Hirayabashi et al. 1985; Masaki et al. 1989; Hirayabashi et al. 1990; Antoniazzi et al. 1991a; and Antoniazzi et al. 1991b). Data from thermal desorption and chemical etching experiments show that the tritium, sorbed on such surfaces after T_2 gas exposure, exists in several forms. Hirayabashi et al. (1985) have indicated that possibly three different species of tritium contamination (one being HTO) are present on stainless-steel samples that have been exposed to high levels of HT gas. Mullins (1992) has reviewed the literature of tritium gas interactions with stainless-steel surfaces and, based on experimental results, generated a model that fits the existing knowledge of tritium on surfaces. He concluded that there is evidence to support the presence of HT, HTO and other unidentified tritiated species on these surfaces. Although there may be tritiated organic species other than HT or HTO, the techniques described here cannot differentiate between them.

The measurement of tritium on surfaces is further complicated by the fact that different methods measure quite different quantities. It is useful to consider at least three aspects of tritium surface contamination. Total tritium includes all the tritium associated with the surface. This value is not particularly useful for radiological-protection purposes. Total tritium is not directly measurable, since destruction of the sample is usually required to release the activity within it. Removable tritium is tritium that can be measured by swipes. Directly measurable tritium is the fraction of the total that is directly on the surface and can be measured with a windowless detector. From a radiation-protection perspective, removable tritium is the most important quantity to measure.

Removable tritium most closely represents the tritium that would be retained by a person who has made contact with a tritium-contaminated surface. Because the tritium betas cannot penetrate the outer dead layer of the skin, fixed-surface contamination (contamination adhering to a surface in such a way that it is not transferable under normal working conditions) is not particularly important, from a radiological-protection viewpoint. The maximum range of tritium betas is about 6 mm in air and about 0.005 mm in tissue. The total tritium surface contamination cannot be accurately determined using any method. Direct methods will not detect all tritium activity below the surface (i.e., "fixed"), due to the short range of the tritium beta particles, and indirect methods generally provide only a reasonable estimate of the "removable" contamination at the time of measurement. Direct surface measurements may overestimate or underestimate the removable fraction by a large factor, depending on the surface finish. Knowledge of the total tritium contamination associated with an item would be beneficial for proper waste management.

The most common indirect method of measuring tritium surface contamination is to swipe the surface, typically 100 cm^2, with a suitable media (ISO 7503-2:1988; Johnson 1991; McElroy and Surette 1992). The swipe is then analyzed for tritium, in a liquid scintillation

counter or a windowless proportional counter. The swipe technique measures the removable tritium and it is estimated, conservatively, that 10% (ISO 7503-2:1988; Surette and McElroy 1988; McElroy and Surette 1992) of the removable tritium is removed by a standard swipe on a previously undisturbed surface. Due to the fact that tritium surface contamination can regrow from the bulk or deep contamination, follow-up measurements should be conducted at regular intervals to verify that the surface contamination has not changed appreciably. It has been shown that stainless-steel planchets exposed to elemental tritium at Chalk River Laboratories (CRL), left undisturbed for a length of time, may lose about 1% of the total tritium per day through regrowth and exchange with the atmosphere (Surette and McElroy 1988). The generic sensitivity of some available instrumentation to measure tritium surface contamination is listed in Table 1.

Table 1: The sensitivity of various survey meters for measuring tritium surface contamination

Detector Type	Sensitivity[1] cm^{-2}	Supplier/Source
Solid-State Detectors	> 30 Bq	Radiation Monitoring Devices - Avalanche Photodiode (direct or smear).
Air Flow Prop. Counter	> 2 Bq	Japanese[2] prototype (direct).
Plastic Scintillator	> 1 Bq	Hughes Whitlock Ltd. Vacuum Scintillation Counter (direct).
Gas Flow Prop. Counter	> 1.5 Bq	Harwell Instruments - Tritium Smear Monitor (smear).
	> 0.4 Bq	Berthold - LB1210D with LB6225 probe (direct).
	> 0.08 Bq	Nuclear Measurement Corp - PC-55 (smear).
Smear/LSC	> 0.02 Bq	Assuming 10% removal (smear).

[1] Assuming a 100 cm^2 area is sampled.
[2] Aoyama et al. 1984; Aoyama and Watanabe 1985.

There can be as many variations in swipe procedures as there are persons doing the swiping. A large variation between different subjects swiping identically exposed planchets and between consecutive swipes by the same person has been noted (McElroy and Surette 1992). All measurements described in this report have been done by one individual. They show a large variation, but are consistent with previous observations.

Although the use of a wetting agent does increase the removal efficiency (McElroy and Surette 1992), it is not recommended. Since the contamination may be absorbed into the structure of the smear material or may be covered by residual moisture or other liquids, the use of a wetting agent may lead to a significant underestimation of the contamination if a proportional counter is used. The range of the tritium betas may be shorter than the liquid film thickness, and the beta particles may never reach the detector. If the smear is counted using liquid scintillation cocktail, the wetting agent must be miscible and compatible with the cocktail used. The use of a wetting agent may further complicate the problem of cross contamination between samples. A highly contaminated wet swipe can easily wet the surface of any shuttle, slide mechanism or gloves, and introduce contamination into the detector or be soaked up by the following samples.

2. EVALUATION

2.1 Liquid Scintillation Counting (LSC)

One recommended method (ISO 1988; Johnson 1991; McElroy and Surette 1992), and the most widely used for tritium surface contamination measurement, is to take a swipe from the surface to be monitored, place it in a vial containing cocktail and count it in a liquid scintillation counter. This method, although very sensitive, is labour-intensive, and the results are usually not available immediately.

The technique used throughout these tests is similar to that described by McElroy and Surette (1992). The use and suitability of various scintillation cocktails and swipe media have been evaluated by Haddock (1992, 1993). Both McElroy and Surette (1992) and Haddock (1992, 1993) concluded that the type of media or the cocktail used was not critical with respect to sensitivity. Other factors affecting the choice of media or cocktail are physical strength, flexibility, price, and cocktail/swipe compatibility.

Ontario Hydro uses a 50 mm (two inch) diameter filter paper for standard smear measurements. These smears generate lint, which may lead to contamination of open window detectors, such as proportional counters. To compare different techniques and instruments, smear/LSC samples were evaluated, whenever possible, on the same surfaces that were used for direct and indirect (smear) measurement with various instruments. Although this "normalization" of measurement was useful, once the contaminated surface has been swiped the contamination conditions will have changed for consecutive swipes. The length of time between swipes also affects the estimate of surface contamination. Not only is the amount of activity in the swipe affected, but the tritiated species may change. This may have some affect on the removal factor and/or the solubility in the cocktail. No convenient method was identified to obtain a standard surface; therefore, direct comparison of tritium surface contamination measurement using swipes was difficult.

Six different smear media were compared: two-inch-diameter Ontario Hydro paper[1]; 25 x 25 mm (1 inch x 1 inch) polyfoam swipes and 4.2 cm ($1^5/_8$ inches) diameter "black swipes", both from ACME[2] ; two-inch-diameter packing foam; and two-inch-diameter discs cut from aluminum foil. Beckman Ready Filters™[3] (glass fibre filters coated with a solid scintillant) were examined in a previous study (Haddock et al. 1993). Although very sensitive, they were not compared to the ones just listed, because they were too fragile.

The methodology used to compare the counting efficiency of these swipe media was similar to that described by Haddock (1992). Ultima Gold scintillation cocktail was used throughout the tests. The foam material tends to partially dissolve in the cocktail and becomes transparent to the scintillation photons, resulting in counting efficiencies comparable to that with just HTO. Results with the pink packing foam indicated a slight quenching in the cocktail, possibly due to discoloration.

The "black smears" are advertised by the manufacturer as being conductive and suitable for low-energy beta counting, such as tritium and ^{14}C. The manufacturer does not indicate from what material these smears are fabricated. This swipe medium discoloured the liquid scintillation cocktail slightly, decreasing the counting efficiency over time. The relative counting efficiencies for swipe media in Ultima Gold liquid scintillation cocktail are shown in Figure 1. These data indicate that there is very little difference between the materials tested.

2.2 Berthold LB1210

For an evaluation of the Berthold LB1210 monitor with both the LB6255 windowless probe and the BZ-200 XK-P Xe counter, see section 2.8.

2.2.1 LB6255 Windowless Detector

The Berthold LB1210D monitor, fitted with an LB6225 windowless gas-flow proportional counter probe, can be used to directly measure tritium surface contamination. The manufacturer's specification states that the monitor with the LB6225 probe can detect 0.37 Bq cm^{-2} (0.1 μCi m^{-2}) of tritium surface contamination (with a background rate of 1 cps) on "relatively large flat areas". The LB6255 probe has a rectangular aperture of 15 mm x 150 mm, with an effective area of 22 cm^2, and uses P10 counting gas.

The Berthold monitor is a well-made, well-designed instrument that can play an important role in the evaluation of surface contamination in a laboratory environment. However, the instrument has no protective window and is susceptible to internal contamination. Its limited dynamic range (three decades) is another drawback. Its background count rate is

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1 Ontario Hydro #016554

2 ACME Distributors, Kingston, Tenn 37763, USA

3 Beckman Instruments, Inc., Fullerton, CA 92634, USA

of the order of 30-50 cps, while its maximum count rate is 3000 cps, an effective dynamic range of only two decades. Therefore, the Berthold LB1210 with the LB6255 windowless detector is not well-suited for routine surveys in a nuclear power generating-station environment.

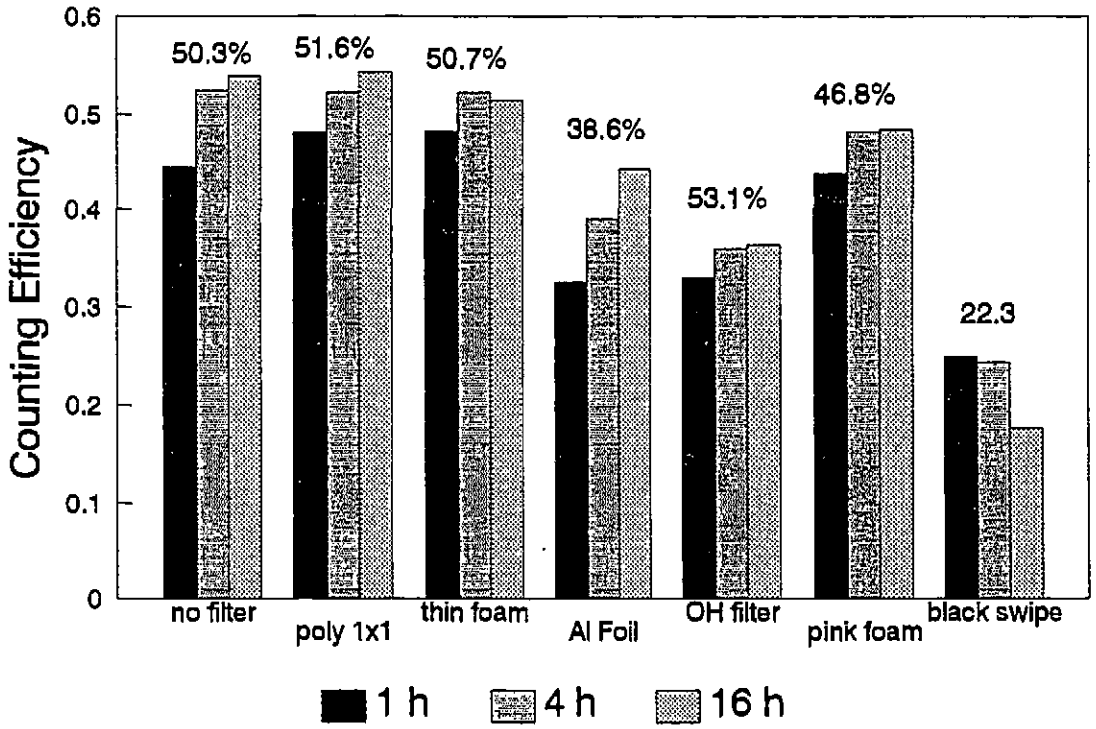

Figure 1: Counting efficiencies of various swipe media in Ultima Gold liquid scintillation cocktail

The Berthold monitor was also tested for measuring swipes directly, with limited success. Although much quicker than liquid scintillation counting and sufficiently sensitive, the Ontario Hydro swipes have fine lint on their surface, due to abrasion. We believe that this contaminated lint was the source of persistent detector contamination.

It is not clear that this monitor measures the same component that is measured with swipes and LSC. Rough surfaces such as absorbent paper or wood make measurement inaccurate, since self-absorption in the contaminated surface can be large, due to the short range of the tritium beta particles.

The LB6255 open-window gas-flow proportional counter, although well-constructed and very sensitive, is easily contaminated with natural oils, dander, and skin flakes that have become contaminated through contact with contaminated surfaces or solutions. When this instrument is used to measure workers' hands or forearms, clothing, or other animals, hair/fibre may protrude into the detection volume and possibly generate false signals by

affecting (static, corona discharge or shorting out the electrode) the electrostatic field inside the detector. The limited dynamic range of the LB1210 also restricts the usefulness of this instrument for routine surveying and monitoring of contaminated surfaces or equipment.

2.2.2 BZ-200 XK-P Xe Counter

The BZ-200 counter is intended to extend the range of the LB1210D monitor. This large-area, sealed, xenon-filled detector has a 5 mg cm^{-2} titanium window. The titanium window is too thick to allow tritium betas to be detected, but the counter could monitor highly contaminated stainless-steel planchets. This is believed to be due to the fact that some of the betas are absorbed in the metal and produce bremsstrahlung, which can penetrate into the detector. The detection of tritium-induced bremsstrahlung typically requires tritium amounts ranging upwards from 50 MBq.

The detector is not sensitive enough to measure tritium contamination on a swipe, but may be useful to measure other contaminants with betas of higher energies, like ^{14}C.

2.3 PC-55 Proportional Counter

The Nuclear Measurement Corporation's (P.O. Box 18248, Indianapolis, Indiana) model PC-55 consists of a 2π windowless proportional counter for measuring both alphas and beta/gammas simultaneously. The monitor is available with detectors of 2.86, 3.49 and 5.7 cm (1, 1.25 and 2.25 inch) diameters. The detector is available either windowless, as tested here, or with a thin (0.25 or 0.85 mg cm^{-2}) mylar window. The monitor has a shuttle mechanism, to allow samples up to 5.4 cm (2.5 inches) diameter to be placed under the detector.

The monitor requires a supply of P-10 (90% argon, 10% methane) counting gas and is conveniently operated by depressing the reset button (after the sample has been placed in the shuttle and located under the detector). The sample can be counted for a preset time ranging from 0.1 to 9999 minutes, or a preset count up to 9999. Whenever a count is performed, the unit automatically purges the detector, with counting gas, for a preset time (OFF, 12 s, 36 s, or 144 s). The results of the measurement are displayed on a seven-decade readout for beta/gamma detection, and on a six-decade readout for alphas. The maximum upper limit of the system is 10^7 CPM. An illuminated bubbler gives a clear indication of the status of the gas flow.

One drawback of this monitor is that it is easily contaminated. Although the swipes are placed on 5 cm (two inch) diameter replaceable stainless-steel sample dishes, to prevent contamination of the shuttle mechanism, the detector was often contaminated after being subjected to samples that had high activity levels. One possible explanation for this is the fact that the detector walls are normally very dry, due to the dry counting gas. Tritiated water vapour from the samples may readily stick to the walls of the detector. With very little humidity to exchange with, the HTO on the walls stays there, causing an elevated background signal. Fortunately, the detector can be easily disassembled, cleaned and reassembled in about ten minutes. Another drawback of this monitor is that there is no

indication that the count rate has been exceeded if a sample with very high activity is tested. If the count rate is exceeded, the detector may be paralyzed and the displays may indicate a low activity, giving a false indication that the surface is clean.

The counting efficiency of the PC-55 was measured using a tritium beta wide-area reference source from Amersham. The beta surface emission rate of the source was 107 s^{-1} ± 10% (1992 July 17) in 2π steradians. The monitor indicated 82 cps, giving a counting efficiency, corrected for decay, of 84%.

To compare the detection efficiency of the PC-55 with swipe/LSC, contaminated surfaces were swiped and the contamination collected was measured using the monitor. After each measurement, the swipe medium was placed in a LSC vial containing ten mL of cocktail and counted. The same six swipe media that were tested for counting efficiency in the LSC were evaluated with the PC-55.

The foam (poly-foam, styrofoam, etc.) showed large variations in measurements on the same surface, even for the same media type. The PC-55 would sometimes indicate large count rates, and at other times it would almost indicate zero. One explanation for this behaviour is that the foam filters are very good electrical insulators and can acquire a static surface charge. This surface charge may be due to either the rubbing action on the surface to be measured, or from the betas on the surface of the media. In any case, the result is that the electrostatic field inside the windowless detector may change shape and adversely affect the collection of ions generated in the detector. Although the foam filters are generally better in cocktail, they are not recommended for use with this type of counter. The apparent counting efficiency observed for the foam filters in the PC-55 proportional counter ranged from less than 1% to more than 10%, compared to that obtained with LSC.

The Ontario Hydro filters were adequate and reproducible, considering that none of the contaminated surfaces are standard. The PC-55 indicated, on average, about 30% of that indicated with the LSC. Figure 2 shows the response of the PC-55 compared with the Packard LSC for Ontario Hydro swipe media from tritium-contaminated stainless-steel planchets. Although the two counting systems use very different processes, they appear to be measuring the same components. The vials containing the swipes were counted after four hours, to allow sufficient time for the activity to leach out from the filter and stabilize using the cocktail.

One volatile component, possibly HTO, of surface contamination that is retained in sealed vials became apparent when a smear was repeatedly counted over a period of many hours. At least two time-dependent components are observed in the plot of activity measured with the PC-55 over time, as shown in Figure 3. The rate of loss of detected activity from the Ontario Hydro filter with a time constant of about 40 minutes may be due to a combination of evaporation and soaking into the bulk of the filter. The longer time constant of more than 30 hours may be due to some stable labelled organic or non-exchangeable tritium slowly being released, as observed by McElroy and Surette (1992).

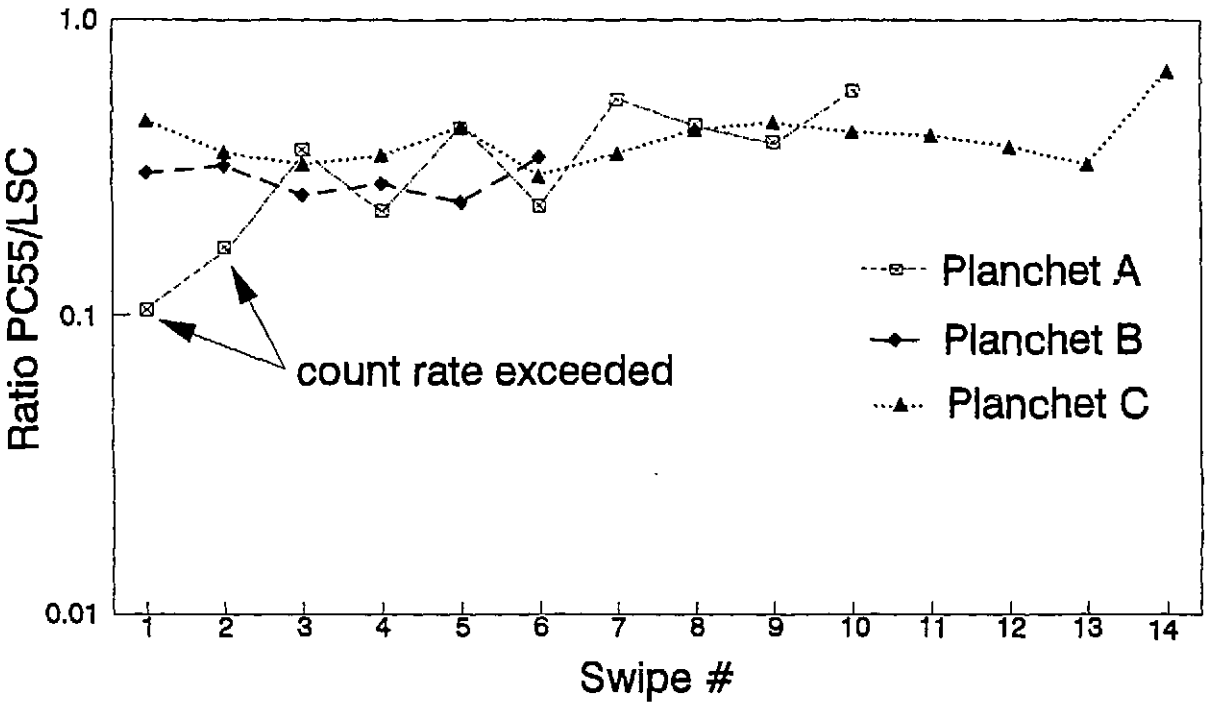

Figure 2: Comparison of the activity measured on Ontario Hydro swipe media using the PC-55 and swipe/LSC

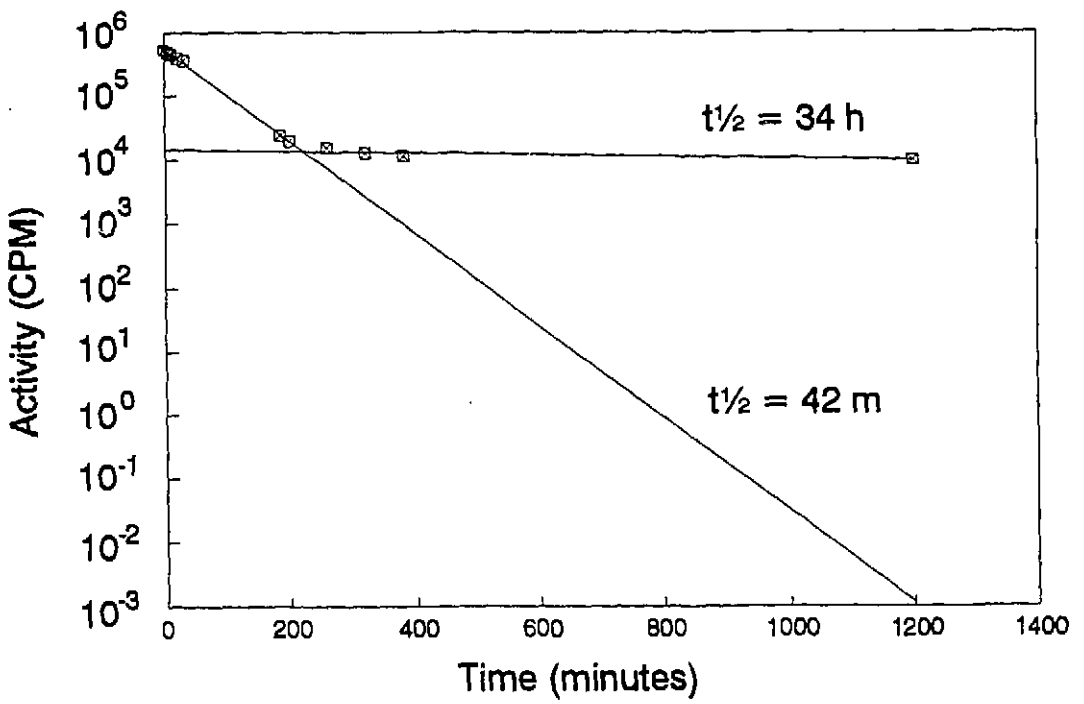

Figure 3: Activity loss, over time, from an Ontario Hydro swipe media measured with the PC-55 proportional counter

The best results obtained with the PC-55 were with aluminum foil and "black swipes" from ACME Distributors. The fact that both of these materials are non-porous and conductive limits the self-absorption and electrostatic effects observed with the other materials. Both media gave results that were comparable with the LSC readings. The "black swipe" readings were generally equal to those measured with the liquid scintillation counter. The PC-55/LSC ratio, with the aluminum foil swipe media, showed a large variation, from unity to more than ten. This is believed to be due to the fact that the non-porous, folded aluminum foil traps most of the activity inside, and a long time is required to allow the activity to equilibrate with the cocktail. The ratio approached unity after a few days.

In summary, the PC-55 is a useful instrument for assessing tritium surface contamination using smears. It has comparable sensitivity and measures the same removable component as LSC measurements. The advantages of having the monitor close to the workplace to be monitored and being able to obtain results immediately more than offset the requirements for a counting gas supply. Th PC-55 has two drawbacks: it seems to be easily contaminated, not only due to lint or dust, but also due to dry detector walls having an affinity to HTO vapour; and, the choice of swipe material is critical. If normal operation with Ontario Hydro filters does not introduce unacceptable contamination, or if frequent decontamination is acceptable, this instrument may fit into operating procedures currently in use at many sites.

The PC-55 is also sensitive to low-energy gamma emitters like ^{55}Fe, and to alpha emitters. It may complement current alpha-beta-gamma monitoring programs. Further operational experience is required to properly assess the usefulness of the PC-55.

2.4 Whitlock Surface Contamination Monitor VSC 5000

The Whitlock Surface Contamination Monitor uses a flat plastic scintillator, 126 mm x 100 mm x 1.6 mm thick, viewed by two 30 mm diameter photomultipliers operating in coincidence, to detect light photon caused by tritium betas. The detector is maintained, via a rubber gasket, at a precise distance (0.5 mm) from the surface to be measured. This gasket also seals the monitor to the surface and prevents external light from interfering with the measurement.

To operate the Whitlock monitor, a partial vacuum (half an atmosphere) is established between it and the surface to be measured. This is performed by actuating a lever that drives a manually operated vacuum pump in the instrument. The light generated in the scintillator is collected by total internal reflection, and counted by a pair of photomultiplier tubes (PMTs) operating in coincidence. Only the disposable gasket is in contact with the contaminated surface. The range of the beta particles in the rarefied atmosphere is longer. This increases the detection efficiency.

Depressing the pump lever releases the vacuum, resets the scaler and allows the instrument to be positioned on the surface to be measured. Releasing the lever provides a vacuum and starts the measurement. The manufacturer specifies a sensitivity of 1 Bq cm^{-2}, over 100 cm^2, for a 10-second count time and assuming no self-absorption.

The Whitlock monitor was evaluated on several smooth flat surfaces and as a swipe monitor. Although the monitor is sensitive, easy to use and moderately rugged, it has severe limitations. It is difficult to find a suitable surface. The manufacturer requires that the surface to be tested be flat to within ±0.125 mm (0.005 inch) over the whole of the detector area of 150 cm^2 (126 x 120 mm). Small scratches and cracks (deeper than 0.125 mm) in the surface to be tested may prevent a vacuum from being established, restricting the operation of the monitor. Porous surfaces such as wood may also prevent a vacuum from being drawn, and since the PMT bias voltage is interlocked with the vacuum, the monitor will not function. Of course, the surface to be tested should be rigid enough to hold a vacuum of up to one half an atmosphere.

The Whitlock monitor can also be used to measure the activity collected on smears. The manufacturer supplies a flat plate with a 50 mm diameter (two inch) recess for the swipe media. The swipe is placed in the depression and the monitor placed onto the plate, covering but not touching the swipe to be measured. The use of a swipe medium with an adhesive back is recommended, to prevent the medium from being drawn, by the vacuum, to the scintillator and contaminating it.

Although the manufacturer does not specify any warm-up time before using the instrument, the initial background readings were usually very high. Figure 4 indicates the time required for the background readings to stabilize after the instrument is turned on. The curve labelled 1st Run represents the time required for the instrument to stabilize after being in storage for more than 24 hours. The other curves (2nd Run and 3rd Run) are representative of warm-up times for the instrument after it was turned off for a brief period (less than 10 minutes). The Whitlock monitor should be turned on for at least one minute before a measurement is attempted.

Another phenomenon observed was that whenever the plastic scintillator was exposed to room lights between measurements, the background readings remained elevated for the first few measurements (20-30 seconds). This may be attributed to the fact that the photo-cathodes on the photomultipliers and/or the plastic scintillator are sensitive to light and may require some time to reach their "dark state" required to measure very low signals.

The monitor is light-sensitive and cannot be used on transparent or semi-transparent surfaces. Some surfaces, like Formica and Melamine, have a transparent finish, which may allow light to reach the plastic scintillator and interfere with the measurement, even if the surface is flat and a vacuum has been achieved.

The advantage of establishing a partial vacuum is that the tritium betas have a greater range and therefore a greater probability of reaching the detector, increasing its sensitivity. Another result of the partial vacuum is that if components of the contamination are volatile, they may change state to a vapour or gas and subsequent readings may be lower. This is indicated in Figure 5, which shows a measurement taken of a swipe from a contaminated surface. Without removing the monitor, a second measurement was initiated. The second measurement was consistently lower than the first. These results indicate that in obtaining the initial measurement, the contamination on the swipe was

altered. Subsequent readings may be due to measurement of a different component and/or mixture of the contamination collected.

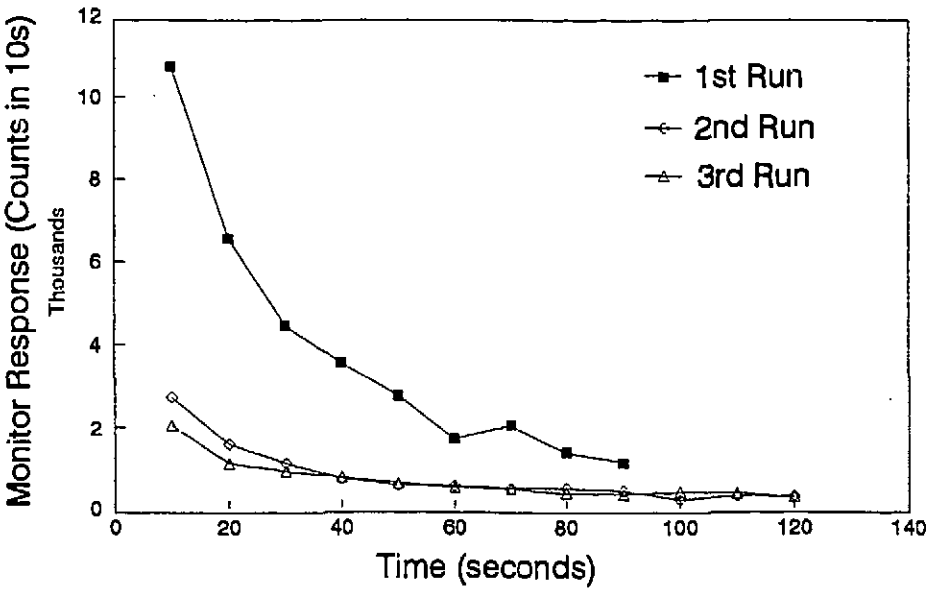

Figure 4: Time required for the Whitlock VSC 5000 surface contamination monitor to stabilize after being turned on

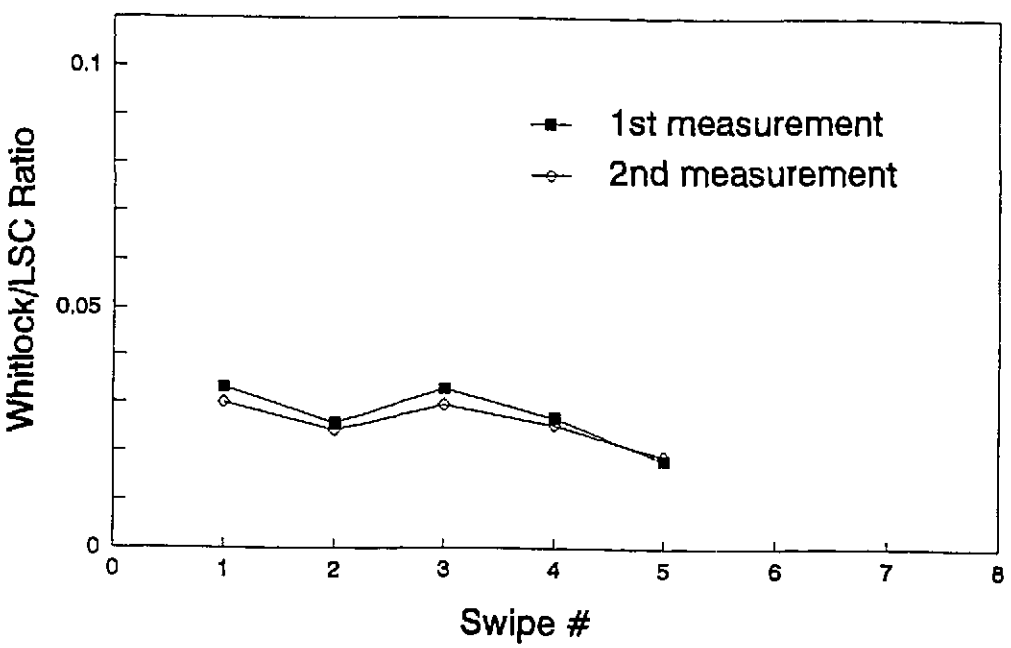

Figure 5: Difference between the first and second measurement for the same swipe (about 20 s between measurements)

Although the Whitlock monitor is very sensitive and easy to use, it has limited use. The requirements of a large, smooth, non-transparent, non-porous and flat surface precludes it from use with most tools and many surfaces. If it is used as a swipe monitor, care must be taken not to scratch the plastic scintillator or contaminate the detector by inadvertently drawing the media to the scintillator with the vacuum.

2.5 MeltiLex™ Melt-on Scintillant

MeltiLex™ is a melt-on scintillant for "dry-on" support counting, for use with the Wallac Filtermat membrane. The manufacturer suggests that MeltiLex™ may be applied without melting (e.g., as an intensifying layer for beta emitters). The application evaluated here is the use of the MeltiLex™ both as a swipe medium and as the scintillant. MeltiLex™ is a wax-based scintillant, durable yet sufficiently flexible to be used in the same manner as the other swipe media tested. It is available in sheets 73 mm wide, 109 mm long, and 0.5 mm thick.

To compare MeltiLex™ with the other methods and techniques tested, two-inch diameter discs were cut from the sheets. These discs were used in the same manner as the other swipe media. Due to the lack of a standard surface, a comparison was made by alternately swiping the same surface with a MeltiLex™ disk and an Ontario Hydro filter paper. It has been reported (McElroy and Surette 1992; Surette and McElroy 1988) that, for a finite number of swipes over a short period of time, subsequent swipes have less activity than the preceding swipes. By swiping alternately with the Ontario Hydro and MeltiLex™ media, it was hoped that a correlation would be observed. Figure 6 shows the relative efficiency of the dry MeltiLex™ compared to the Ontario Hydro medium in Ultima Gold. The MeltiLex™ swipes were placed in empty 20 mL LSC vials while the Ontario Hydro swipes were placed in vials, containing 10 mL of Ultima Gold liquid scintillation cocktail. The Ontario Hydro medium appears to remove a larger, and possibly different, component than the MeltiLex™. The MeltiLex™ results were very reproducible and stable; virtually no time was required for the signal to stabilize.

No significant difference was noted in the activity measured for MeltiLex™ swipe with respect to the orientation of the material in the counting vials, indicating that the scintillation photons are not absorbed in the material. This was determined by comparing the activity measured using swipes that were left open and free-standing in the vial to the activity measured using swipes that were folded twice, to one quarter of their original size, with the contamination on the inside, and that were placed on the bottom of the vial.

MeltiLex™ swipes were also tested in a Nuclear Enterprise[4] (NE LSF-1) single vial counter, modified to accept the MeltiLex™ disks without the need of a vial. The counter uses two PMT's in coincidence mode, and could easily be modified to accept a shuttle mechanism for MeltiLex™ disks. The advantage of this setup is that there is no need for sample preparation, cocktail or vials, and the results are available immediately. The results using

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[4] Nuclear Enterprise Inc., 931 Terminal Way, San Carlos, CA 94070, USA

this prototype were similar to those using vials in an LSC counter, but with lower efficiency. No attempt was made to optimize the prototype NE counter.

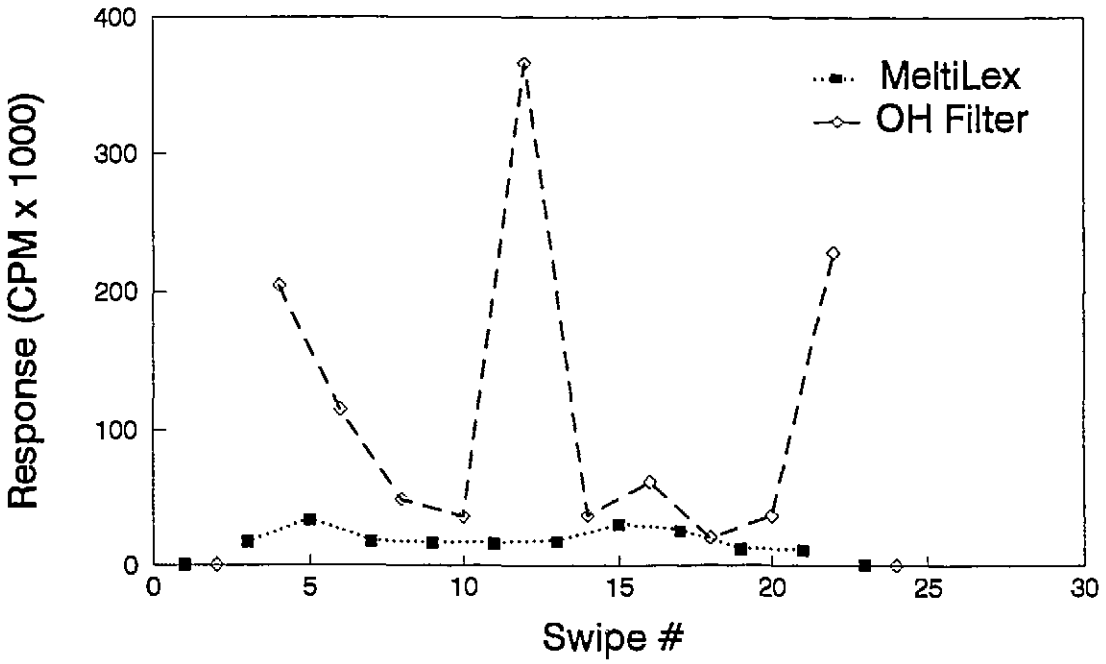

Figure 6: Response of MeltiLex scintillant as a swipe media compared to the activity measured with an Ontario Hydro swipe in Ultima Gold liquid scintillation cocktail. The same surface was alternately swiped with each media. Swipes #1, 2, 23 and 24 are background samples.

To compare the removal efficiency, the MeltiLex™ disks were placed in the PC-55 windowless proportional counter. It was assumed that only the surface contamination collected on the disk would contribute to the signal, and not the scintillations due to betas in the MeltiLex™. The activity measured using the PC-55 was much lower than that observed by measuring the scintillations in the MeltiLex™. It is believed that, on swiping, the surface of the plastic flows, covering the contamination or embedding it into the scintillant. This will actually enhance the production of fluorescence, due to the betas, but will reduce the number of particles emanating from the surface and reduce the signal measured by the proportional counter. This hypothesis may also explain the stability of the LSC/PMT measurements over time. The contamination may be trapped into the MeltiLex™, giving it intimate contact with the scintillant, and preventing the contamination from leaching out.

A disadvantage of using MeltiLex™ is that the plastic is hydrophobic and may not give a proper indication of the surface contamination if there is a large HTO component. An attempt was made to measure small quantities of HTO applied directly onto the plastic, with little success. In addition, although the material was sufficiently durable to use as swipes once cut into disks, some of the sheets in the package were shattered. MeltiLex™ becomes brittle when cooled. The package may have been dropped while frozen (it was delivered in December) and some of the pieces may have broke at that time.

The MeltiLex™ wax-based scintillator may be useful for routine tritium surface contamination measurements. Further tests are required to characterize its removal efficiency, species dependency and gamma interference. It may be possible to discriminate between tritium and other contaminants collected on the smear by pulse height or rise time discrimination. If energy discrimination is possible, MeltiLex™ may be suitable for alpha/beta surface contamination determination. A prototype monitor may not be very sensitive to external gamma interference, because the material is so thin and less susceptible to internal contamination, since the scintillant is replaced after each measurement.

2.6 Electret Surface Monitor

E-perm™ electret ion chambers, adapted for surface contamination measurements, were evaluated. The prototype *E-perm*™ ion chambers were supplied by Biomation,[5] a distributor for Rad Elect Inc.[6] The prototype electret tritium surface contamination monitor consists of a small-volume (~20 cm^3) ion chamber with a 38 mm (1.5 inch) diameter window. A measurement is taken by removing the protective "keeper" from the electret, installing the surface monitor window and placing the chamber with the open window on the surface to be measured. The activity of the measured surface is determined from the difference in the electrostatic potential on the electret surface before and after the measurement, and the time of exposure to the contaminated surface. A calibration factor, for each unit, is determined experimentally.

The manufacturer quotes a calculated sensitivity of 1.67 V h^{-1} for a contamination level of 1000 dpm cm^{-2} (~4.5 μCi m^{-2}). Although these are sufficiently sensitive to measure well below the 18.5 MBq m^{-2} (500 μCi m^{-2}) derived surface contamination limit for surfaces exposed to HT (Whillans 1986), they may not be sensitive enough for routine surface contamination measurements; about two hours is required to measure tritium contamination at a level of 5 μCi m^{-2}.

2.7 Herfurth "Microcont" Surface Monitor

The Herfurth "Microcont HGZ 190" is a surface-contamination monitor that has a large-area flow proportional detector for alpha/beta measurement, with a built-in gas supply. Intermittent flushing of the detector with the built-in butane/propane gas is required. The gas capacity is 75 cm^3 and can be monitored with a level gauge that is viewed through a sight glass. A single filling of the tank is sufficient for one week. An accessory for this monitor that was not evaluated, because it was not supplied, is a sample slide mechanism for positioning swipe samples or aerosol filters under the detector for reproducible counting geometry.

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[5] Biomation, 335 Perth St., Almonte, Ontario, Canada

[6] Rad Elect Inc., 7499 Whitepine Road, Chesterfield Business Park, Richmond, Va, USA

The effective window size 150 cm^2 and 0.7 mg cm^{-2} thick, which may allow some of the more energetic tritium betas to reach the detector and be detected. The manufacturer quotes an efficiency of 15% for ^{14}C. Due to the thin window on the detector, it was possible to measure the activity collected on an Ontario Hydro smear, although the relative efficiency is about 10^5 less than swipe/LSC measurements. It was also possible to directly observe the difference in the surface contamination of a stainless-steel planchet before and after swiping the surface with an Ontario Hydro filter. The contamination levels on the planchets used were much higher than would be expected in a normal workplace environment. Figure 7 shows the relationship between swipes measured with the Microcont and the LSC, and also direct measurements of the contaminated planchet. The activity measured on the swipes with the Microcont followed the same trend as the LSC measurements, but at a much lower efficiency. The reduction in surface contamination due to that removed with the swipe is apparent in the direct surface measurement using the Microcont.

The Microcont is much more sensitive to other radioisotopes than for tritium, and is not suited for routine tritium contamination measurements. However, it may be a useful instrument for other low-energy beta-gamma emitters, like ^{14}C and ^{55}Fe. The Microcont is not sensitive for skin measurements, nor suitable for measuring small, uneven surfaces.

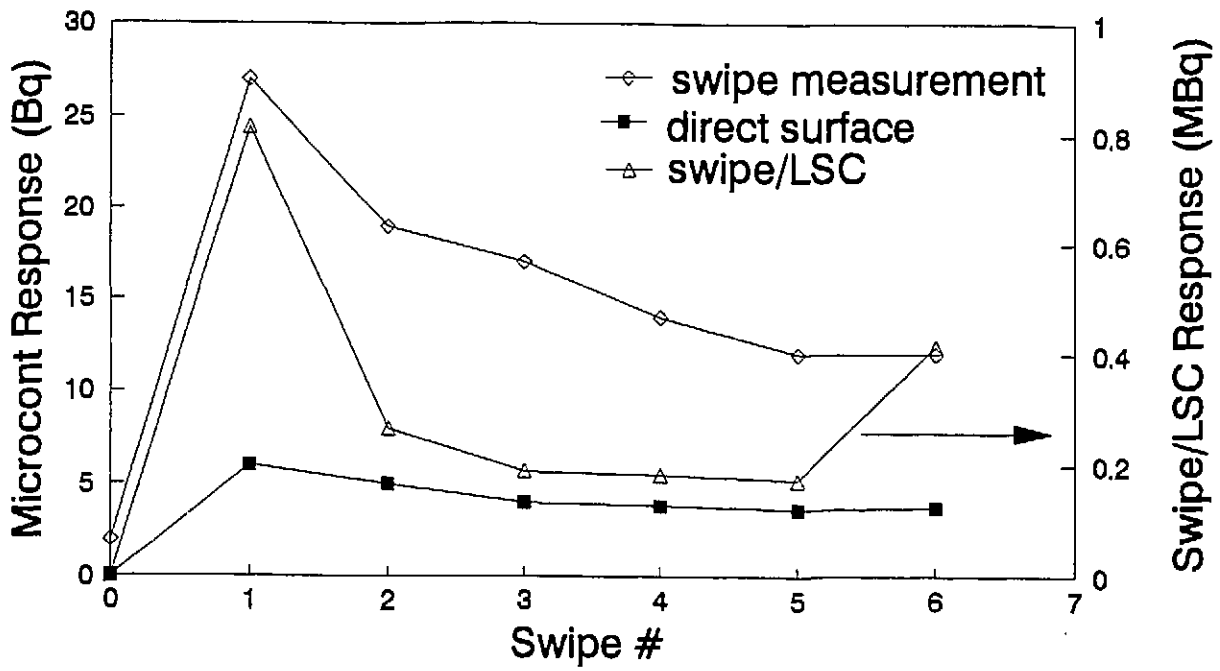

Figure 7: Comparison on Herfurth Microcont Monitor with LSC measurements (direct and indirect)

2.8 Skin Measurements

2.8.1 Direct

Hairless rats exposed to about 1 MBq of tritium from both HT-exposed stainless-steel planchets (Trivedi 1993; Trivedi et al. 1989) and tritiated oil (Trivedi 1992) were monitored using the LB6255 windowless proportional counter probe. The probe was fitted with an aperture plate to restrict the size of the measuring area to 1.8 cm^2. The manufacturer recommends using the aperture plate when measuring surfaces that are not flat or that do not fully cover the window opening. It is also useful for monitoring fingertips and pinpointing the location of contamination on surfaces. The area on the hairless rats that was exposed with either the oil or the stainless-steel planchet was in the order of 10 cm^2.

Due to the limited dynamic range of the LB6255 probe, the results are not quantitative. In general, immediately after exposure, the measured activity at the point of contact for both the tritiated oil and tritiated stainless-steel planchet was more than 3000 cps (over the instrument's range). The activity measured, immediately after exposure, on an unexposed part of the rats was at background levels. The exposure location was on the left rear flank of the rats exposed to the stainless-steel planchets, and on the back of the neck for the rats exposed to the tritiated oil.

On the day following the exposures, the rats were again measured at the exposed location and at an unexposed site. The activity measured at the exposure site for both the tritiated oil and stainless-steel planchet exceeded the upper range of the monitor (>3000 cps). The rats exposed to the contaminated surface did not show any contamination above background on the unexposed skin, but the rats exposed to tritiated oil on the back of their necks had activity levels that were about twice background on unexposed skin. This may be due to the low vapour pressure of the oil and the fact that the rats regularly groom themselves. The oil does not evaporate readily and the rats can spread the contamination to other parts of their bodies. However, the contaminated area (the back of neck) is difficult for the rats to reach.

Three hairless rats were also contaminated with 10 kBq of tritiated oil on the back of their necks as described above. Unfortunately, the measurements on the 1 MBq exposures had contaminated the detector such that the background was now elevated to >150 cps, compared to ~50 cps. The rats were not monitored immediately after administering the contaminated oil, but on the following day. The activity detected at the point of exposure was about 100 cps above background, and no activity above background levels was detected on an unexposed area. Although it was possible to measure surface contamination due to contact with tritium-contaminated surfaces and tritium-contaminated oil, immediately after exposure and at least one day after exposure, an accurate measure of the activity level was not possible. The calibration factor for the probe with the 1.8 cm^2 aperture is not given, the counting geometry is not fixed, and the self-absorption of the tritium betas in the skin is unknown. In addition, the behaviour of tritium on skin is such that, after even a few minutes, some of the tritium would have soaked into the skin and been undetectable to external monitoring.

The BZ-200 XK-P Xe probe was evaluated with hairless rats exposed to about 1 MBq of tritium from tritium-contaminated stainless-steel and 1 MBq of tritiated oil. In both cases, no activity was measured at the point of contact immediately after exposure. No attempt was made to measure the unexposed areas or to measure the rats one day after the exposure.

The Whitlock VSC 5000, the electret surface monitor and the Herfurth Microcont are not suitable for direct measurement of skin contamination. The Whitlock VSC 5000 requires a larger surface than is normally found, and skin does not support a vacuum. The Herfurth Microcont registered a reading only at the higher contamination levels, and the electret-based prototype requires a long sampling time.

2.8.2 Indirect/surrogate

The direct measurement of tritium-contaminated skin was done on rats that were used in dosimetric experiments (Trivedi 1993; Trivedi 1992; Trivedi et al. 1989). Swipes could not be taken from these rats as it would have interfered with the uptake and subsequent analysis of the excretion of the tritium. Due to the complexity and cost associated with breeding rats, and uncertainties in the activity on the contaminated surfaces, a substitute was sought.

Skin is a complex organ, made up of a cutaneous layer that includes a "dead layer" and a basal cell layer, and a subcutaneous layer that includes fat, sebaceous glands, hair follicles, and muscles. Pig skin is very similar to human skin, and possibly hairless rat skin, with respect to structure, and may serve as a mimic for surface-contamination monitor evaluation.

Johnson et al. (1988) have shown that tritium in hairless rat skin more than 250 μm deep contains less than 50% of the total activity after four hours and only increases a moderate amount after 24 hours.

To evaluate the suitability of using fresh pig skin for surface contamination evaluation, a section of it was subjected to contact exposure using a tritium-contaminated stainless-steel planchet. The section of skin was divided in two. After four hours post-exposure, one section was cut into thin slices using a cryo-cooled microtome. The tissue slices were solubilized and the activity measured. The remaining section of skin was treated in the same manner 24 hours after it had been exposed with the planchet. For direct comparison with Johnson et al. (1988), the activity in successive 250 μm layers was determined (0-250, 250-500, 500-750, and 750-1000 μm). The results from the depth profile of the pig skin indicate that the total activity transported past the 250 μm layer is about 30% that measured in live rat skin. This observation was consistent for both the four-hour and 24-hour elapsed times after exposure.

Several pieces of pork flesh were exposed, using the same protocol described by Trivedi (1993); i.e., a contaminated planchet was applied with a pressure of 44 g cm^{-2} for one minute. The skin surface contamination was then measured with consecutive swipes. The total activity transferred to the skin was determined to be 3 MBq. The activity

removed by the first swipe was about 16% of the total, while the activity removed by five consecutive swipes and by washing was about 80% of the total activity transferred to the skin sample. This is consistent with observations by Johnson et al. (1988). The removal efficiency, as determined by the slope of a line fitted to the swipe data of the Ontario Hydro swipe media on the pig skin tested, is 10% per swipe.

Although the active transport mechanism (blood capillaries) is not available in dead, fresh pig skin, we may assume, from this test, that over a short period of time the tritium sink due to fat and tissue will help the contamination diffuse into the flesh. From these data, we have made the assumption that pig skin is a reasonable model for human skin contamination transport at the point of contact, for the first few hours after exposure.

Counting the activity collected on swipes from contaminated skin with the PC-55 and swipe/LSC was the only effective evaluation method. The Whitlock VSC 5000, although sufficiently sensitive, was not convenient, because it required an adhesive-backed swipe and was easily contaminated. The wax-based scintillant MeltiLex™ was not evaluated as a method of measuring tritium skin contamination.

3. RECOMMENDATIONS FOR FUTURE WORK

Other than the obvious follow-up operational experience with the PC-55, to determine whether it is suitable for routine swipe analysis, and to determine the availability, durability and cost of a MeltiLex™ based prototype monitor, one type of monitor developed for alpha detection warrants further investigation.

A Long Range Alpha Detector (LRAD) (MacArthur et al. 1992; MacArthur et al. 1993a; MacArthur et al. 1993b), can satisfactorily monitor irregular surfaces or inside spaces smaller than the probe's dimensions. It is also suitable for monitoring large surfaces, and can be easily automated. The mechanism for detection is to transport the ionized molecules, generated by the contamination, by a moving air current into a flow-through detector. A "multi-grid" ion chamber is currently used to monitor the ion current generated. The LRAD is most sensitive to alpha particles.

Because the LRAD is sensitive to ions rather than the alpha particles directly, any radiation that can create ions, including neutrons, beta particles and gamma rays, may be detected. Low-energy betas, such as tritium, which have a maximum range, in air, of less than a centimetre, may be readily detected, because the ionized molecules can be transported by a moving air current into a flow-through ion detector.

Garner et al. (1993) have predicted, using Monte Carlo calculations, that the LRAD is about three orders of magnitude more sensitive to a 5 MeV alpha emitter than it is to tritium. Based on these calculations, the LRAD technology could be used as a tritium surface monitor with sufficient sensitivity for current regulatory limits. The ion lifetime in air has been measured to be about five seconds (MacArthur et al. 1992). If an air flow of 5 L min^{-1} is used and one assumes that all ions are collected, then the signal current generated is similar to that of an ion chamber with a volume of about 400 mL.

A monitor designed using the LRAD technique could be used to monitor the hands and arms of radiation workers for low-energy beta emitters, including tritium. Any object (tools, parts, electronic equipment, clothing, etc.), including hands and arms that have potentially been contaminated, could be monitored on the entire outer and inner surfaces. Contaminated surfaces that are accessible to the air flow will produce ions. Thus all surfaces will be monitored simultaneously. Even small cracks and corners can be effectively monitored if the air passes over them.

Although the LRAD provides no information as to the nature of the contaminant, it may be useful as a screening device. Once an object has been evaluated for gross contamination using conventional survey meters, the LRAD could be used to determine whether the object is free of low-range radiation, such as tritium, ^{14}C or possibly ^{55}Fe.

4. CONCLUSIONS

Although a number of instruments can detect tritium from contaminated surfaces, none are more sensitive than the swipe-LSC counting method. The PC-55 monitor can be as sensitive as the swipe/LSC method with the proper choice of swipe media. Both the PC-55 and LSC measure the quantity of interest (i.e., the removable surface contamination). The PC-55 offers the convenience of obtaining the result right away, and can be located close to the workplace to be monitored, making it much more convenient to use. One drawback of the PC-55 is the fact that it can be readily contaminated internally. With care, the detector can easily be decontaminated. It is felt that this monitor can serve a useful purpose in an effective radiation-protection program.

The limited dynamic range, and susceptibility to contamination, of the Berthold LB1210 monitor with an LB6255 windowless probe restricts its usefulness to relatively clean laboratory environments. The Berthold monitor equipped with a BZ-200 XK-P probe is not sufficiently sensitive for routine tritium surface-contamination measurements. The Berthold surface-contamination monitor may be useful in a relatively clean and controlled environment.

The usefulness of the Whitlock VSC 5000 is limited, due to its requirement for a large, smooth, flat, non-porous and non-transparent surface. The VSC 5000 is very sensitive and may be useful for measuring swipes. One advantage is that the VSC 5000 can be easily transported to a different workplace, and swipes are easily measured with a backing plate equipped with a recess for the swipe media. Its requirement for an adhesive-backed swipe media and possible contamination may limit its usefulness in the workplace.

The Herfurth Microcont is easy to use and, although not sensitive enough for routine workplace surface monitoring, it may play a valued role in areas where high contamination are expected, such as in tritium handling facilities. The electret-based prototype surface-contamination monitor requires too long a sampling time for low-level tritium surface-contamination measurements.

A monitor using MeltiLex scintillant may offer a convenient alternative to LSC counting. The MeltiLex scintillant is adequately sensitive for routine low-level surface-contamination measurements, and the technology exists to fabricate a suitable swipe monitor. The availability, durability, and cost of the MeltiLex sheets would have to be investigated before a swipe monitor based on this medium could be recommended for routine use.

Preliminary investigations suggest that fresh pig skin may be used to simulate the kinetics of human skin for the first few hours after contact with tritium-contaminated surfaces.

It is recommended that the PC-55 windowless proportional counter be used in real field conditions, to assess whether internal contamination problems will limit its usefulness for routine measurements. If it is determined to be unsuitable for routine surface-contamination analysis, then a MeltiLex-based monitor should be pursued. The possible use of LRAD technology for surface tritium contamination monitoring also warrants further investigation.

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