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Title: STUDY OF NASAL SWIPE ANALYSIS METHODS AT LOS ALAMOS NATIONAL LABORATORY

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DISCLAIMER

STUDY OF NASAL SWIPE ANALYSIS METHODS AT LOS ALAMOS NATIONAL LABORATORY

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Abstract-**The Health Physics Analysis Laboratory (HPAL) performs around 30,000 nasal swipe analyses for transuranic nuclides each year in support of worker health and safety at the Los Alamos National Laboratory (LANL). The analysis method used employs cotton swabs swiped inside a nostril and liquid scintillation analyses of the swabs. The technical basis of this method was developed at LANL and has been in use for over 10 years. Recently, questions regarding the usefulness of a non-homogenous mixture in liquid scintillation analyses have created a need for re-evaluation of the method. A study of the validity of the method shows it provides reliable, stable, and useful data as an indicator of personnel contamination. The study has also provided insight into the underlying process which occur to allow the analysis. Further review of this process has shown that similar results can be obtained with different sample matrices, using less material than the current analysis method. This reduction can save HPAL the cost of materials as well as greatly reduce the waste created.**

Key words: nasal swipe; liquid scintillation; plutonium; transuranics; radiation work

INTRODUCTION

Nasal swipe analyses have been recommended for use in personnel protection from radionuclides, in particular transuranics, through inhalation [Faust, 4.51; Volex, 50; Ritch, 6-2]. In the past, nasal swipes have been performed using a paper filter or cotton wrapped around a stick. The swipe material was then removed and analyzed using a gas proportional counter or hand held α instrumentation [Faust, 4.45; Volex, 50]. The gas proportional method was reported to have a loss of 25% of the activity due to depth of burial [Ritch, 6-84] as well as a 50% loss due to geometry if a 2π counter was used (or an increase in time to analyze the sample on each side). This method is further complicated by the need to dry the swipe and manipulate it into the gas proportional counter if α contamination is to be seen. It has also been reported that liquid scintillation analysis can be used on nasal swipe analyses, and this method alleviates the need for drying the swipe [Ritch, 6-84].

The technical basis of the nasal swipe analysis method used at LANL is provided in an internal LANL document, HPAL-TA-55, "Quality Assurance Manual, Plutonium Liquid Scintillation Methods and Procedures," prepared by Romero. This document provides comparisons of different swipe media,

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cotton swabs and Whatman-40 paper, different counting methods, gas proportional and liquid scintillation, and different chemical compositions of Pu nitrates and oxides. These comparisons showed that liquid scintillation provided good efficiencies and simpler analyses, so it was adopted for use at LANL. Also, using a counting range optimized for α particles, the study showed cotton swabs and Whatman-40 filter papers provided similar results, so cotton was adopted as the sample of choice because of its ease of use. This method has been in use by LANL under this technical basis for approximately ten years.

The Health Physics Analysis Laboratory (HPAL) is the team specifically charged with these analyses and performs around 30,000 each year. The cotton swipes are moistened and swiped around the inside of a worker's nostril (one swipe is used for each nostril). The swipes are delivered to HPAL in a box which contains the two swipes from each individual. They are prepared for analysis by clipping the cotton head off the swab into a 20 ml liquid scintillation vial. Ten (10) ml of scintillation fluor is added to the vial, the cap is attached, and the vial is agitated for approximately 5 seconds. The vial is placed into a liquid scintillation counter and allowed to dark adapt for at least 20 minutes to remove transient luminescence before it is analyzed. This procedure is consistent with the aforementioned methods developed at LANL.

Liquid scintillation records the light pulses produced by the scintillation fluor which has been excited by the absorption of ionizing radiation. The signal produced by each light pulse varies depending upon the type and energy of the radiation. Pulses of various amplitudes can be separated, and an energy spectrum produced. HPAL uses two main regions of interest in the energy spectrum for all analyses. A 0-25 keV region (Region A) region is used primarily to detect β^- particles, specifically ^3H (maximum β^- energy 18.6 keV, unquenched), and encompass the full range of the ^{241}Pu β^- particle (maximum β^- energy 21 keV, unquenched). A region from 25-800 keV (Region B) is used primarily for α particle detection. In liquid scintillation, the optimal analysis for α particles is 100% efficient and the energy reported is approximately one tenth of the actual energy (e.g., a 5 MeV α will produce a peak near 500 keV on the energy spectrum). Nasal swipe analyses at LANL are performed to detect transuranic radionuclides which have α energies ranging between 4 to 6 MeV. The number of counts which appear in Region B is of principal interest in nasal swipe analyses (typically the spectrum is not reviewed).

Questions have arisen as to the effectiveness of the current method of nasal swipe analysis, which have caused HPAL to undertake the study described in this paper. The optimal conditions for liquid scintillation include a homogenous mixture of the substance being analyzed and scintillation fluor. This is not the case for the current method of nasal swipe analyses as the tip of cotton swab, usually resting at the bottom of the vial, is included in the sample matrix. Although it is of utmost importance that a reliable analysis is developed, other considerations such as cost and materials must be taken into

account. Furthermore, many of the nasal swipes analyzed at HPAL are "Emergency" swipes taken as a result of airborne radiation limits being exceeded in an area where unprotected workers are present. It is therefore important that the nasal swipe analysis process provide results as quickly as possible. Using these criteria, the study revealed three major components of nasal swipe analyses; determining how the non-homogeneous matrix affects the results (Validation of Analysis Method), optimizing the sample matrix (Modifications to Analysis Method), and evaluating the effect of the β^- particle from ^{241}Pu found in transuranic mixtures at LANL (The ^{241}Pu Component).

EXPERIMENTAL

Validation of Analysis Method

The cotton tips of the wood swipes used at LANL were spiked with known amounts of different transuranic α emitters (multiple analyses for ^{241}Am, ^{239}Pu, and ^{238}Pu were performed). The tips were spiked from a pipette with a 100 µl of solution, which was diluted to provide an activity of approximately 450 pCi (1000 dpm) on the swipe. One hundred (100) µl was chosen to completely saturate the tip without dripping, providing for equal deposition of the solution throughout the swab. The swabs were allowed to dry, and then analyzed using the current method for nasal swipes. Nitrate solutions were used on all the spiked cotton swabs. Nitrate solutions were chosen because they are most common in Pu work at LANL. Furthermore, oxides are usually created by exposure of Pu to air and thus can be avoided by careful working procedures [Romero; Faust, 1.8].

The data from the analysis showed that Region B, where the α activity from α particles should be collected, varied between 85- 95% of the actual value. It was further determined, by the method of least squares, that these values remained constant and stable from the first analysis (approximately 30 minutes from the start of preparation) to analyses several days later. Review of the spectra show a vastly degraded α peak which spreads from the actual α energy all the way down to 0 keV, as shown in Figure 1.

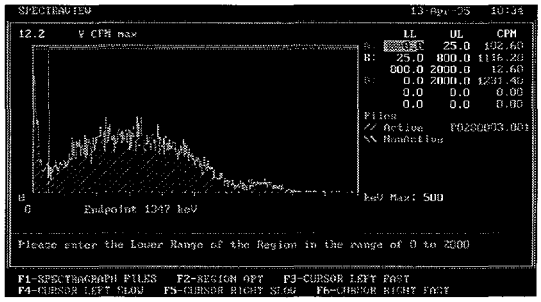

Figure 1 Example of degraded spectra from ^{239}Pu on a cotton swab

Removing the swab from the original vial and fluor and placing it in a new vial with new fluor showed that the vast majority of the activity remained on the swab.

The degraded spectra is assumed to be caused by attenuation of the α particles by the cotton swab. Further experiments were conducted by adding water to the vial in hopes of removing the activity from the swab by solubilizing the nitric solution, and suspending it in the fluor (a major part of scintillation fluor is an emollient designed to suspend liquids, the fluor itself does not suspend solids). This

would create a "pseudo-homogenous" environment in the fluor not immediately around the swab [This may not be the case for oxides as they are much less soluble in water than nitrates (Wick, Taube)]. It was observed that the water does suspend the particles into the fluor and thus resolves the α peak, as seen in Figure 2.

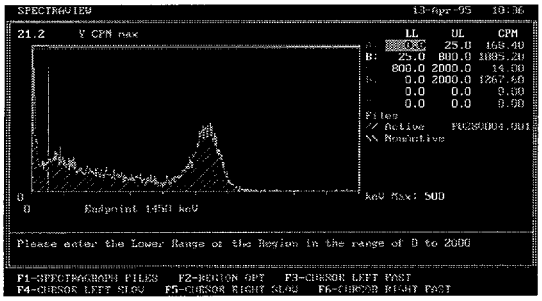

Figure 2 ^{239}Pu spectra of the nasal swipe from Figure 1 with 0.5 ml H_2O added.

However, as found by Romero and duplicated in this study, this process was very gradual over time, as seen in Figure 3.

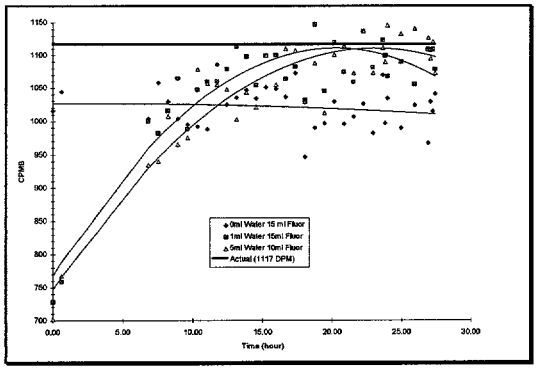

Figure 3 Activity seen for ^{238}Pu in various analysis matrices with 2nd order curve fit.

Figure 3 shows that when the samples with water were analyzed immediately after sample preparation, only 50 to 60% of the activity was seen. Over time, 10 to 15 hours, the α peak became resolved for these samples, and values fluctuating around 100% of the actual activity was seen. Romero reports resolution of the α peak in 50-60 hours using water with Insta-Gel, a Toluene based fluor. HPAL currently uses Packard Ultima-Gold, a slower, "environmentally friendly" fluor). However, as previously mentioned and illustrated in Figure 3, the samples without water added displayed very stable results from the first analysis to analyses several days later, but they fluctuate around a value lower than the actual value. In hopes of expediting the gradual in-growth of activity in the samples with water, different aliquots of water, intense agitation of the swab with water only and with a water and fluor mix, and heating the water, was tried, but no observable change from the slow in-growth was seen.

Modifications to Analysis Method

Because of the behavior of the analysis method seen in the validation stage of the study it was determined that other modifications to the current method might be possible. From the previous analyses it was determined that the majority of the activity being seen by liquid scintillation was remaining on the cotton swab. It was therefore speculated that only the fluor which was immediately near the swab was necessary to record the activity, and the majority of the 10 ml of fluor was not serving a useful purpose. All liquid scintillation equipment in use by HPAL is capable of handling 7 ml "Pony" scintillation vials. A comparison of a spiked swipe in a Pony vial, with 2 ml of fluor (sufficient to cover the swipe) and the analysis matrix currently in use by HPAL of 20 ml vial and 10 ml flour, was

conducted. A drawing of the 2 different sample matrices is shown in Figure 4.

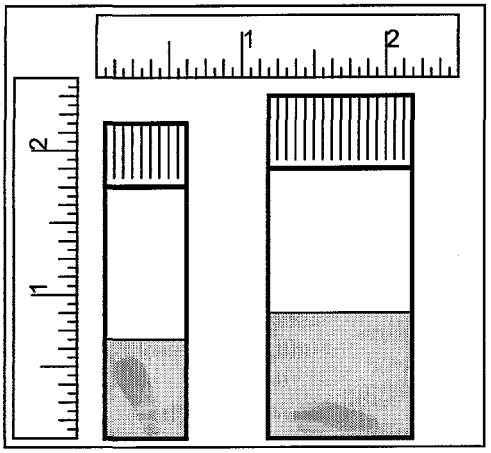

Figure 4 Nasal swipe analysis matrices.

Liquid scintillation analysis of the two different sample matrices for various transuranic α emitting nuclides shows that the activities reported are, in all cases, similar. Furthermore, comparing the spectra from the 20 ml vial (see Figure 5) and the Pony vial (see Figure 6) shows them to be remarkably similar (multiple analyses of ^{238}Pu, ^{241}Am, and ^{235}U were performed).

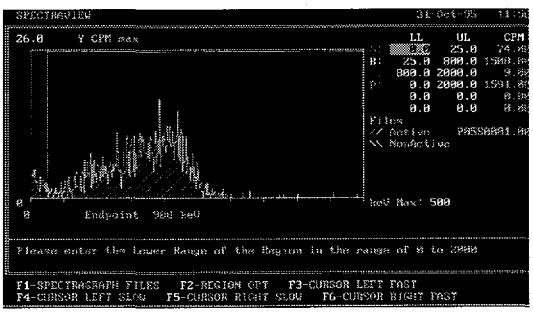

Figure 5 ^{235}U spectra from 20 ml liquid scintillation vial with 20 ml fluor (no water).

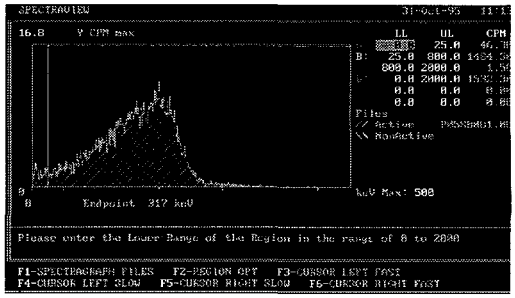

Figure 6 ^{235}U spectra from Pony liquid scintillation vial with 3 ml fluor (no water).

The ^{241}Pu Component

A ^{241}Pu β^- particle is expected to be seen, in Region A of most positive nasal swipes, due to the Pu material mixtures commonly handled at LANL. In fact, it is usually highest in specific activity of all radionuclides in the mixtures. ^{241}Pu contributes to the number of counts seen in Region A of the spectra and, therefore, combined with the information obtained in Region B from α particles, it should be useful in the analysis method or as a method of analysis by itself. To explore this possibility, several samples were prepared by spiking ^{241}Pu on a swipe in the same method as discussed previously. It was observed that the counting efficiency for the ^{241}Pu β^- particle, which has a similar energy to ^{3}H, is much less than expected (the correct efficiency, of approximately 50%, is seen for ^{3}H) and erratic for samples prepared in the same manner.

RESULTS AND DISCUSSION

Validation of Analysis Method

It was concluded that the current method of nasal swipe analysis, although not optimal, is adequate to meet the

necessary analysis requirements. Although the analysis does not report 100% of the actual activity, the stability of the method is better suited to the required conditions of a quick and simple analysis. Furthermore, the detection limits of liquid scintillation analyses are sufficiently low, when compared to the limits determined by worker health and safety, that even with the 5 to 15% loss of activity, transuranic radionuclides are still easily detected. Due to the time requirements of nasal swipe analyses, adding water to resolve the α spectrum and provide 100% efficient collection is not feasible[1].

Modifications to Analysis Method

The study of the LANL nasal swipe analyses method provides information into the nature of the underling mechanism of the analysis matrix. The data indicate that the activity, and thus the radionuclide particles in the presence of scintillation flour, remain in the cotton swab. It is concluded that the analysis matrix need only include a sufficient amount of fluor to cover the swab. A smaller analysis matrix and less fluor can be used. The optimization of the analysis method would provide savings in both material and waste.

The ^{241}Pu Component

It was also concluded that the data collected from ^{241}Pu, which contributes to the overall spectra and thus counts collected in Region A, is not reliable because it is not efficient or stable[2]. Therefore, its contribution attributed to ^{241}Pu should be removed from the analysis. This is currently accomplished by eliminating the counts seen in Region A. Plutonium-241 can still be seen in the sample spectrum (and in fact its β^- peak provides further proof that any positive swipes are "real" and not the effect of some other luminescence) and used in the analysis, but characterization of the sample activity from the ^{241}Pu component is not reliable.

Recommendation

It is the recommended that nasal swipe analyses performed by HPAL be optimized by using the following sample matrix:

- The tip of the cotton nasal swipe
- 2-3 ml of scintillation fluor
- 7 ml pony vials

This recommendation does not further complicate the current analysis method.

FUTURE WORK

HPAL will continue to conduct further studies in nasal swipe analysis in hope of

[1] Transuranics, and in particular isotopes of Pu, have an affinity of sticking to solids. It is known that strong acid leaching (normally a combination of nitric, hydrochloric and hydrofluoric acids) is required to solubilize Pu [Cook]. However, adding acid to nasal swipe analyses is not an option at HPAL due to waste concerns, and was not considered.

[2] Cook and Anderson provide an explanation for the erratic ^{241}Pu results in their paper "The Determination of ^{241}Pu by Liquid Scintillation Spectrometry Using the Packard 2250CA". They explain that using the burst counting circuit on Packard liquid scintillators (one of Packard's methods to reduce background counts by analyzing and discarding signals which have "after pulses") and a slow, "environmentally safe", fluor such as Ultima Gold, eliminates many of the actual ^{241}Pu counts.

finding an analysis method which will fully resolve the α spectrum and report 100% of the actual activity while not adding to the analysis time or materials. Studies to include the addition of different, non-hazardous, materials to the matrix, tests of different liquid and solid paraffin fluors, and the possibility of obtaining scintillation machines which can accommodate smaller sample matrices and provide a further reduction in the use of vial material and fluor are planned.

A further study of the apparent discrimination of ^{241}Pu β^- particles is also planned. This study is of interest for two very different reasons. First, to help in nasal swipe analyses performed at HPAL, a method to optimize the machine settings to provide useful information from the ^{241}Pu β^- could provide better analysis results. Also, on a broader scale of analyses, it would be interesting to determine the underlying mechanism which causes the elimination of ^{241}Pu β^- particles to occur. This phenomena is not seen for the ^{3}H β^- which is very similar in energy to the ^{241}Pu β. Apparently, Packard liquid scintillation machines can, to some extent, discriminate between very similar β^- particles based upon the parent nuclide.

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