RAPID MONITORING FOR TRANSURANIC CONTAMINATION DURING BURIED WASTE RETRIEVAL*

G. G. Loomis
C. V. McIsaac
Idaho National Engineering Laboratory
EG&G Idaho, Inc.
Idaho Falls, Idaho

ABSTRACT

Research on rapid "on line" measurement techniques for monitoring the presence of transuranic (TRU) contaminants during retrieval of buried defense-related transuranic waste was conducted at the Idaho National Engineering Laboratory. Multiple techniques for measuring both loose and airborne contamination were evaluated in actual dusty, contaminated conditions. The contaminants were isotopes of plutonium and ^{241}Am.

For airborne contamination four different continuous air monitors were evaluated including the KURZ 8311 System,** the RADeCO 452 System, an ANL-W system, and the Victoreen 758. For loose contamination, six different techniques were examined including alpha liquid scintillation using the photon-electron rejection alpha liquid scintillation system, rapid radiochemical separation followed by alpha spectroscopy, gamma spectroscopy for the ^{241}Am 59 kev gamma, x-ray spectroscopy for the uranium L-shell x-ray, direct gross alpha counting for inference of the presence of plutonium isotopes, and finally, direct alpha counting using a large area detector.

These monitoring devices were experimentally evaluated for sensitivity in pico-curie per gram; pico currie per sample, or pico curie per cubic centimeter and rapidity of turnaround time for an individual analysis. Levels of loose contamination can be measured at 1 to 100 pCi/g at the 95% confidence level with about a 1-hour turnaround. For airborne activity, all four continuous air monitors met or exceeded 0.002 pCi/L in 8 hours.

INTRODUCTION

This report presents the results of research covering rapid "on line" monitoring for the presence of transuranic (TRU) contaminants during retrieval of buried defense-related TRU waste. The main contaminants to be monitored were compounds of plutonium and americium. Rapid "on line" monitoring for both airborne and loose contaminants is part of an overall contamination control strategy involving various misting and fixant sprays to prevent the spread of dust associated with retrieval operations and will be used to track trends of the airborne and loose contamination levels within the retrieval operation.

A series of techniques for measuring both airborne and loose surface contamination have been identified and experimentally evaluated for potential use in an integrated contamination control system. For airborne contamination, several state-of-the-art several alpha continuous air monitors (CAMs) were evaluated for sensitivity to contaminants and rapidity of measurement. For loose contamination, measurement techniques include alpha liquid scintillation, alpha-radiochemistry techniques, gamma spectroscopy for americium gammas, x-ray spectroscopy for ^{239}Pu presence from the uranium L-shell x-ray, direct filter counting for gross alpha, and direct counting with spectroscopy for ^{239}Pu analysis.

This report will first discuss background information about buried TRU waste and the contamination control system of which the rapid monitoring system is a key component. A description of the expected contamination will be presented. Second, the overall concept for obtaining trend level for monitoring strategy is given. Third, the results of research on air monitors and loose contamination monitoring are presented. Finally, recommendations for which strategies should be utilized in a rapid monitoring system are given.

BACKGROUND/EXPECTED CONTAMINANTS

Between 1952 and 1970, over 65,000 m^3 of TRU mixed waste, which was primarily generated at Rocky Flats Plant (RFP), was stored at Idaho National Engineering Laboratory (INEL) in shallow land-filled pits and trenches. During 1987, organic solvents such as trichlorethyne and carbon tetrachloride were found in the aquifer 500-600 ft below the buried waste, and trace amounts of plutonium were also

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present at 110-ft inner bed.* Because of these positive identifications of migration, retrieval of the waste for processing and final disposal is being considered.

During FY-89, INEL performed conceptual and preliminary designs for a remotely-operated retrieval system that could be safely used in a hot demonstration retrieval of a buried pit at INEL. The overall retrieval system design involves containment buildings, remotely-controlled excavators and transporter, separation systems, a contamination control system, and final disposal options.

The contamination control system was based on the concepts of maintaining the natural moisture content in the pits and trenches to reduce the fugitive dust during excavation, a laminar flow ventilation system to control inner-building air quality, and effective monitoring of contamination spread.(1) The philosophy of the contamination control system was to maintain a relatively radiologically clean operation allowing rapid personnel entry to perform routine and nonroutine maintenance on the remotely operated equipment.

The contamination control system design involved a Dust Suppression System (DSS) and a Rapid Monitoring System (RMS).(2) The DSS is a grouping of subsystems including an inner-building laminar flow ventilation system. The remotely controlled Lifting and Moving System (LAMS) provides mobility for the Contamination Suppression System (CSS). The CSS is a series of misting/fixant sprays and vacuum systems for dust control.

The RMS will provide "on-line" monitoring of airborne and loose plutonium/americium contamination within the inner-building. In addition, the techniques developed for this inner-building monitoring are applicable to other areas of retrieval including the separation/processing and storage systems. By maintaining "on-line" monitoring of airborne and loose contamination, established safety and operating limits can be followed. These limits for entry into contaminated areas with bubble-suit protection are conservatively set by the Department of Energy set at a limit 10,000 times the maximum permissible concentration limit (unprotected access). As retrieval operations proceed, examining the ongoing levels of airborne and loose contamination trends, the retrieval techniques, and the contamination suppression techniques can be altered to meet the limits. This strategy of a relatively low operating limit and an on-line knowledge of the trend of contamination levels will expedite the overall retrieval process by reducing downtime for elaborate cleanup. The INEL philosophy on safety demands "no uptake" of plutonium, and these low limits support this philosophy.

The contaminants to be monitored are oxides, chlorides, nitrates, and hydroxides of ^{238}Pu, ^{239}Pu, ^{240}Pu, and ^{241}Am. Because ^{241}Pu (also present in the waste) has a relatively short half-life and decays by beta emission to ^{241}Am, americium will always be present with the plutonium. The primary source of the INEL waste is from manufacturing processes at RFP where the americium impurity was processed out to varying degrees with various waste streams. As part of this process, the americium was concentrated in resin columns that were sent to INEL as waste. The ratio of americium to plutonium by activity for most waste streams averages about 7.4.(3) However, this ratio varies considerably depending on waste stream; therefore, monitoring activities require both plutonium and americium monitoring.

Extensive plutonium recovery operations were conducted at RFP, and therefore, only extremely fine particles of plutonium/americium interspersed in salts and absorbent reached the INEL waste pits. For example, of the tons of plutonium processed by RFP, approximately 360 kg of ^{239}Pu (the primary isotope by mass) was sent to INEL. This 360 kg is interspersed with 2×106 ft^3 of waste and 6×10^6 ft^3 of fill dirt.(4) On a TRU waste definition basis, the average retrieved waste would be below the 100 nCi/g limit on ^{239}Pu alone; however, the relatively small mass of high specific activity americium caused retrieved waste to average 140 nCi/g.

RFP plutonium/americium particles are primarily 0.3 - 3 μm diameter particles that are expected to electrostatically** attach to larger negatively charged dust particles associated with the fill dirt.*** Dust particles greater than 20 μm AMD [aerodynamic mean diameter] will quickly fallout within a few feet of suspension (5); therefore, most of the dust containing plutonium to be monitored by the RMS will be less than 20 μm AMD.

OVERVIEW OF MONITORING SYSTEM

During a retrieval operation, the RMS will be displayed to give hourly contamination level status for all areas associated with retrieval including the retrieval inner-building connection tunnel, separation area, and processing area.

* Communication from T.H. Smith (EG&G) to T. R. Gessel (DOE-ID) on subsurface investigation results, June 15, 1987.

** When an alpha particle (+2 charge) leaves the nucleus of the plutonium, electrons can be stripped leaving the plutonium/americium particles positively charged. The plutonium/americium particles is a conglomeration of hundreds of thousands of atoms of plutonium/americium and decay products.

*** Private communication with Gehart Langer, Rocky Flat Plant, Golden Colorado.

For airborne activity monitoring, multiple (up to six) alpha CAMs will be located on movable tripods throughout the retrieval area, and on-line readings will be processed through a central PC-based computer system where hourly readings will be stored and displayed. Operational and safety alarm limits will activate a standard alarm system.

For loose contamination, both fallout coupons and smears will be processed continuously during routine, manned bubble-suited entry (such as for maintenance and repair). The fallout coupons will be remotely collected on a routine basis using the LAMS and transporter system.(3) The fallout coupons will be dispersed and collected throughout the retrieval area using a special canister holder by LAMS and attached to and moved by the transporter to a glovebox port for manual transfer to an adjacent counting laboratory. Data from the loose contamination counting schemes will be logged into a PC-based data system that will be coordinated with the airborne alpha CAMs output and continuously printed with hourly updates.

Data will be continuously examined for changes in level of airborne and loose activity. As mentioned previously, INEL safety will allow bubble-suited entry at airborne levels below 10,000 times the maximum permitted concentration of plutonium in air or loose contamination; however, to allow a clean and uninterrupted retrieval, a fraction of that level will be allowed . The RMS will be required to rapidly track extremely low levels of contamination (1 to 100 pCi/g for filters and smears and 0.002 pCi/L for air) from multiple samples such that retrieval techniques can be quickly altered if an observed trend shows an unacceptable change in contamination levels (even in a strong radon/thorium backgrounds).

A brief description of both airborne and loose contamination monitoring schemes and their measured sensitivities follows. Details are given in Reference 6 about the various monitoring schemes.

AIRBORNE CONTAMINATION: ALPHA CAMS

The four alpha-CAMs experimentally evaluated in this study (KURZ 8311; RADeCo 452; Victoreen 758; ANL-W System) were all shown to meet or exceed the DOE order 5480.11 requirement for measurement sensitivity (0.002 Pci/L in 8 hours.). Three of the CAMs were evaluated in a dusty environment of aerosolized soil spiked with ^{239}Pu (the KURZ, RADeCo and Victoreen), and the ANL-W system was evaluated in a dusty environment with clean soil only. The soil was aerosolized by a dust generator consisting of a trough with tygon tubing containing small holes emersed in the soil. Compressed air forced dust particles into the air. The soil was spiked with ^{239}Pu to 8 nCi/g.

Because of high radon and thoron daughter backgrounds at INEL, alpha CAMs with high sensitivity to measure the presence of specific isotopes (^{239}Pu) must efficiently account for the naturally occurring alpha emitters, even though the alpha spectrums are of similar energy. To accomplish this, three different algrothms were used, referred to here as the two-, three- or four-window alogrithm.

$$Cp_u239 = C_1 - k_1C_3 \quad \textit{Two-window} \qquad \text{(Eq. 1)}$$

$$Cp_u239 = C_4 - k_2 (C_3 + C_5) \quad \textit{Three-window} \qquad \text{(Eq. 2)}$$

$$Cp_u239 = C_6 - k_3 \frac{C_7C_8}{C_9} \quad \textit{Four-window} \qquad \text{(Eq. 3)}$$

Where C_1, C_4, and C_6 are the total spectral counts under the ^{239}Pu peak, and C_2, C_3, C_5, C_7, C_8, and C_9 are the total number of counts under the corresponding background peaks. The constants K_1, K_2, and K_3 are experimentally evaluated in a dusty environment involving clean dirt.

TABLE I

^{239}Pu LLD Concentration (pCi/L)

CAM	Two-windows	Three-windows	Four-windows
30 minute counting interval:			
KURZ	6.0 ± 0.2 E(-04)	5.2 ± 0.2 E(-03)	1.7 ± 0.2 E(-03)
RADeCO	1.01 ± 0.09E(-02)	1.12 ± 0.04E(-02)	8.9 ± 4.3 E(-03)
Victoreen	1.2 ± 0.1 E(-02)	1.45 ± 0.06E(-02)	7.8 ± 0.2 E(-03)
60 minute counting interval:			
KURZ	2.19 ± 0.05E(-04)	1.8 ± 0.2 E(-03)	6.1 ± 0.5 E(-04)
RADeCO	3.8 ± 0.4 E(-03)	1.29 ± 0.07E(-03)	4.1 ± 2.1 E(-03)
Victoreen	4.4 ± 0.4 E(-03)	5.2 ± 0.2 E(-03)	3.0 ± 0.8 E(-03)
ANL-W[a]	2.1 ± 0.2 E(-04)	2.2 ± 0.2 E(-04)	1.5 ± 0.1 E(-04)

a.Performed in clean aerosolized dirt only.

These ^{239}Pu counts were then converted using detector efficiency to pCi and correlated to the amount of air drawn through the CAM head to get pCi/L sensitivity.

Table I summarizes the experimental results, giving the sensitivity as a lower limit of detection (LLD) in pCi/L. These LLDs were determined using the method of Currie.(7)

Of the three alogrothms used, the four-window approach displays consistently lower LLDs than the two- or three-window approach. Although not tested in plutonium contaminated dirt, the ANL-W system appears to result in generally lower LLDs. All CAMs displayed excellent sensitivity for application to retrieval of buried TRU waste, and all display LLDs below DOE Order 5480.11 (0.002 pCi/L in 8 hours.). The generally lower LLDs for the 60-minute count compared to the 30-minute count are due to improved statistics using the Currie method for determining LLDs.

LOOSE CONTAMINATION

Six different rapid and accurate techniques for measuring the presence of loose TRU contamination fallout filters, smears, or direct soil samples were evaluated. The sensitivity of these techniques varied between extremely fine (1 pCi/g in 1 hour) to fine (100 pCi/g in 1 hour). The six techniques included rapid radiochemistry and spectrometry, alpha liquid scintillation, gamma counting for ^{241}Am, x-ray counting for U-L shell x-rays, gross alpha counting, and finally, direct counting using a large area spectrometer. For all these techniques, blanks and TRU contaminated samples were prepared to evaluate the sensitivity and turnaround time. The samples were prepared using existing radiochemistry techniques.(4) LLDs were evaluated using the Currie technique. A brief description of the techniques and a summary of their sensitivity and turnaround follows.

Alpha Liquid Scintillation

Alpha liquid scintillation involves a fairly complex chemical dissolution aided by microwave digestion. The resulting "cocktail" is a scintillant liquid that contains the TRU particles. The resulting light pulse from emission of alpha particles is counted and correlated by standard spectrometry. The main application for this technique is for fallout coupons and filter smears or CAM filters. The proton-electron rejection alpha liquid scintillation system was used for this analysis.

Rapid Radiochemistry and Spectrometry

This method involves complicated chemical dissolution, depositing the solutions into a membrane filter and counting alpha particles with a surface barrier detector and spectroscopy. This technique gives the most sensitive results for soils samples, smears, and air dust on filters.

Gamma Counting for ^{241}Am

This technique involves standard gamma spectroscopy using Germanium detectors to count the 59 kev gamma of ^{241}Am always present with defense related TRU waste. Due to a large variation in the amount of americium, this scheme can be used to assess quantitive presence of americium only.

U-L Shell X-rays

This technique involves utilizing the decay of ^{239}Pu to U and involves analysis of the L-shell x-ray emission from uranium. Large area Germanium spectrometers with thin sample holders are used for direct counting.

Gross Alpha Counting

This technique involves direct alpha counting of contaminated soils or filters on a plate with a large area scintillation detector. No spectroscopy is involved; however, background alpha interference from ^{233}U and ^{232}Th daughters must be well-understood. Also, allowance for the decay of shorter lived Ra daughters must be allowed to decay between counting intervals. This technique does not quantify the presence of TRUs but only gives a qualitative indication of a presence above background, which is useful for screening purposes.

Large Area Direct Counting with Spectroscopy

This technique counts alpha directly--with essentially no preparation--with a large area, custom-built gridded ionization chamber (Ordella Inc., Oak Ridge Tennessee, Model 8200A). The diameter of the chamber is 35 cm, and the diameter of the sample holder is 24 cm.

Summary of Sensitivity and Turnaround for Loose Contamination

Table II summarizes the measured sensitivities along with expected turnaround times for the loose contamination measurment systems. For the most sensitive measurements, but not necessarily the most rapid, radiochemistry and alpha liquid scintillation should be applied. For fine sensitivity and rapid turnaround, U-L shell x-ray and gross alpha techniques can be applied. For determining the presence of ^{241}Am, gamma counting for the 59 kev gammas applies. The large area direct spectroscopy has potential to provide both high sensitivity and rapid turnaround. The high sensitivity is attributed to the large area sample plate and large volume detector, and rapid turnaround is attributed to the short and simple sample preparation.

CONCLUSION

A variety of schemes for both loose and airborne TRU contamination have been evaluated and shown to be applicable to handling of TRU wastes in retrieval operations. A

TABLE II
Loose Monitoring Techniques.

METHOD	SAMPLE SIZE	SAMPLE PREPARATION	COUNT TIME	BACKGROUND (cpm)a	LOWER LIMIT OF DETECTION	TURNAROUND TIME
Alpha liquid scintillation	10 mg - 1 g	Microwave digestion + 2 liq-liq. extractions	10 to 30 min	0.2	1 to 5 pCi/g	60 min for 1, 2 hours for 6
Rapid radiochemistry	>1 g	Fusion followed by chemical separation	15 min	0.005	~1.4 pCi/g	70 min for 1, 260 min for 8: 10 min more for dusts
241AM gamma counting	10 mg - 1 g	none	60 min	2 to 20	10 - 30 pCib	~ 60 min
L-shell x-ray	~ 6 g	none	60 min		< 100 pCi/g	~ 60 min
Gross alpha	~ 1 g	Prevention of added exp. to air, or allow overnight decay of radon	30 min		~ 16 pCi/g	30 min. to hours depending on sample decay
Direct alpha spectrometry	0.25 g	Grinding and spray deposition on large St. steel pan	15 min	~0.5	~ 40 pCi/g	~ 45 min

a. counts per minute (cpm).

b. Sample mass must be known to convert to pCi/g (i.e., if 10 pCi is detected in a 10 mg sample, then the LLD is 1000pCi/g).

combination of these techniques can provide an "on line" trend of levels of contamination within a retrieval area such that safety and operating limits are seldom challenged.

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