Implementation of a Surface Contaminated Object Characterization Program at LANL – 14346

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ABSTRACT

Radiological characterization is an important component to disposition and disposal of radioactive waste from Los Alamos National Laboratory (LANL). One characterization technique that has been historically used and continues to be used at LANL includes gamma and neutron based non-destructive assay. This technique is generally very effective, however, for waste items that are surface contaminated it is beneficial to introduce an additional characterization approach based on direct surface surveys. This characterization approach for surface contaminated objects (SCO) has been employed at other DOE sites including Rocky Flats, Hanford, and Oak Ridge. NRC/DOT NUREG 1608/RAMREG-003, “Categorizing and Transporting Low Specific Activity Materials and Surface Contaminated Objects” is the driving guidance document for the radiological characterization of surface contaminated objects. This paper describes the SCO program in effect at LANL as adapted from the Rocky Flats and Hanford programs.

INTRODUCTION

A surface contaminated object (SCO) is a solid object which itself is not radioactive, but which has fixed and/or removable radioactive contamination distributed on any of its surfaces. NRC/DOT NUREG 1608/RAMREG-003, “Categorizing and Transporting Low Specific Activity Materials and Surface Contaminated Objects”[1], gives guidance on how to prepare both low specific activity (LSA) and SCO radioactive waste for shipment in accordance with Federal regulations – 10 CFR 71, “Packaging and Transportation of Radioactive Materials”[2], and 49 CFR 173, “Shippers General Requirements for Shipments and Packaging”[3]. In addition to complying with NRC/DOT NUREG 1608/RAMREG-003, Department of Energy (DOE) sites must meet the requirements of DOE O 435.1, “Radioactive Waste Management”[4] and its accompanying manual DOE M 435.1-1. Guidance on meeting LSA and SCO requirements has recently been published by the DOE as DOE-STD-5507-2013, “Standard for Communicating Waste Characterization and DOT Hazard Classification Requirements for Low Specific Activity Materials and Surface Contaminated Objects”[5]. In addition, all radioactive waste must meet the disposal facility’s waste acceptance criteria prior to shipping for disposal.

The Los Alamos National Laboratory (LANL) SCO program was developed to provide the radiological characterization for currently generated and legacy surface contaminated process waste - this includes a number of gloveboxes, process hoods, pencil tanks and similar items from the LANL Plutonium Facility, Radioactive Liquid Waste Processing Facility, the Chemical and Metallurgy Research Building, and the decommissioned plutonium processing lines at Technical Area 21 dating from LANL’s early plutonium operations. Since development, the SCO program has expanded to include other types of radiological waste such as shielding and equipment used in depleted uranium operations.

SCO characterization is based on sampling the contamination level per area on a surface. Therefore, essential components of the program are: an accurate calculation of the surface area, reliable health physics instruments, representative sampling points, and a defensible statistical calculation of the level of activity for an SCO item or group of items. Lastly, for items contaminated with transuranic (TRU) radionuclides, an accurate waste mass is required to determine whether the items can be disposed of as low-level waste. Oversized items that do not meet the low-level waste criterion are either size reduced for disposal as TRU waste or decontaminated to less than 100 nCi/g of transuranics.

SCO DEFINITION

The regulatory definition of SCO can be found in 49 CFR 173.403, "Surface Contaminated Object (SCO) means a solid object which itself is not radioactive but which has Class 7 (radioactive) material distributed on any of its surfaces. SCO must be in one of two groups with surface activity not exceeding the following limits:.."

The definition goes on to state the limits for two categories of SCO using activity per surface area levels for SCO I (lower activity) and SCO II (higher activity) in activity per surface area units. There are two separate activity type limits – one for beta, gamma and low toxicity alpha emitters and another for all other alpha emitters. There are separate limits for fixed and non-fixed (removable) contamination and accessible surfaces have a much lower limit than inaccessible surfaces. The regulatory SCO I and SCO II upper limits in units of activity per unit area are detailed below in Table 1.

Table 1 SCO Upper Limits

Units	Bq/cm2		µCi/cm2	
Category	Alpha (Bq/cm^2)	Beta/Gamma and Low Toxicity Alpha (Bq/cm^2)	Alpha ($\mu Ci/cm^2$)	Beta/Gamma and Low Toxicity Alpha ($\mu Ci/cm^2$)
SCO-I				
Accessible, non-fixed (removable) contamination	< 0.4	< 4	< 0.00001	< 0.0001
Accessible, fixed contamination	< 4,000	< 40,000	< 0.1	< 1
Inaccessible, non-fixed and fixed contamination	< 4,000	< 40,000	< 0.1	< 1
SCO-II				
Accessible, non-fixed (removable) contamination	< 40	< 400	< 0.001	< 0.01
Accessible, fixed contamination	< 80,000	< 800,000	< 2	< 20
Inaccessible, non-fixed and fixed contamination	< 80,000	< 800,000	< 2	< 20

The distinction between the type of radiation and between fixed and non-fixed contamination is straightforward, but the accessible/inaccessible determination requires some judgement. An accessible surface area is one that could be contacted if the packaging is removed by an accident

– for purposes of measuring the contamination, the rule of thumb is that any surface which can be readily reached by a person's hand, using standard radiation measuring techniques is an accessible surface. Any other surface is called an inaccessible surface. This can be further defined as any surface which requires disassembly to be surveyed using standard radiation measurement techniques or requires a tool to isolate or gain access to an area. For example, the inside of a pipe might be inaccessible past the first few inches. Of course, there still must be a means of determining the activity level of inaccessible surfaces to assure that the applicable SCO limit is met. This can be done by acceptable knowledge or by measurement. Accessible surfaces can also be made inaccessible after measurement, e.g., installing a threaded cap, welded/bolted/strapped flange/plate, injection of foam or grout, or application of fixative coatings may be used.

In addition to meeting the contamination limits, the waste must also meet other transportation requirements. For example, the waste must meet the definition of non fissile or fissile excepted material, the external radiation limits, and total activity limits. The container type is determined by the level of activity of the waste.

THE LANL SCO PROGRAM

Development of the LANL SCO program started with a request from the LANL Plutonium Facility to use SCO to characterize several gloveboxes that were in the process of decontamination and decommissioning from process lines with an eye to disposing of the gloveboxes as low-level waste. Attempts had been made to characterize the gloveboxes in place using non-destructive assay, however, given that in-process radioactive material was present near the gloveboxes, this had proved unproductive. The decontamination process itself required that the gloveboxes remain attached to HEPA filtered facility exhaust systems until the process was complete. A site specific program needed to be developed to handle this waste stream. It was determined fairly early on that several DOE sites had fully developed programs in place to take advantage of this characterization method. In particular, we were able to work with current or former personnel from the Rocky Flats Environmental Technology Site (RFETS) and the Plutonium Finishing Plant at the Hanford Site [6,7] to develop the LANL program. The natural follow-on to the initial development effort was to expand the program to include other waste that qualified as surface contaminated radioactive waste.

The process starts with identifying a waste item or group of items with the same process history that qualify as surface contaminated. Having a similar process history suggests that a group of waste items has comparable isotopic distribution and levels of contamination. Being surface contaminated seems relatively easy to ascertain in most cases, obviously including sheet metal, piping, gloveboxes, rigid plastics, laboratory cabinets, and flooring, and just as obviously leaving out activated materials, sludges, filter media, insulation, and other debris type waste. Other items such as demolished process building material, painted surfaces, and contaminated items with some permeable and some impermeable surfaces have to be assessed on a case by case basis. The requirements for categorizing a waste as SCO allow having a nominal amount of LSA material mixed in with SCO waste.

Once it is ascertained that a waste qualifies as a surface contaminated waste, the next question to answer is whether the isotopic identification and distribution of radionuclides is known or can be determined. This is necessary because the survey instruments used to characterize the level of alpha or beta/gamma contamination of the surfaces measure disintegrations per minute per surface area, they do not identify the radionuclides. At LANL, this isotopic distribution is determined in one of several ways. For the legacy waste, where acceptable knowledge may be inadequate, far field gamma spectroscopy can be used to determine the detectable isotopic distribution. The undetected but expected radionuclides can be inferred using LANL specific isotopic distributions of standard DOE material types. If needed, smear samples can be taken and sent for isotopic analysis in a laboratory. For newly generated waste, the material type is known, and may be supplemented by radiological characterization of smear samples to determine exact ratios of radionuclides of interest, for example, to determine the ratio of americium to plutonium in a glovebox being decommissioned.

The next question to answer is whether the waste can be adequately characterized by a quantitative survey of the surfaces. LANL utilizes statistical methods developed at RFETS to determine what surveys are necessary to provide adequate characterization. An object can be adequately characterized using one of two statistical methods (Plan A or B) or a non-statistical method (Plan C). Plan A is a conservative statistical method intended for use when it is known that contamination levels do not closely approach the DOT SCO limits. Plan A employs non-parametric analysis and produces results that are sufficiently accurate to ensure compliance with shipping regulations and waste acceptance criteria. Plan B is especially intended for use when contamination levels approach the DOT SCO limits or disposal facility waste acceptance criteria (WAC) limits, but could be used in any activity scenario. When this alternate statistical method is selected, the calculations of total activity and activity concentration in the shipping container use a mean estimator of surface contamination level. Plan B is less conservative, yet continues to provide high confidence that the material meets the criteria for shipment under the appropriate SCO classification. Plan C is a non-statistical method for characterization when the entire surface of the object(s) is measured, similar to performing a release survey. Since the data is comprehensive, statistical tests are not necessary. The measured maximum values are used for comparison to SCO limits and the mean value is used to calculate the package specific activity. Plans A and C have been the primary methodologies used in the LANL SCO program. For Plan A characterization, LANL has developed a standard protocol of requiring 30 or more survey points for both inaccessible (e.g., inside a glovebox) and accessible (e.g., outside a glovebox) areas of a waste item. Spreadsheet utilities have been developed and are used for the required statistical calculations. These spreadsheets automatically check to determine whether SCO limits are met or exceeded and alert the analyst of an exceedence.

The total surface area of an SCO is required to determine the total activity of the item and is obtained either by direct dimension measurements or by estimation methods allowed by the LANL SCO procedure. An accurate waste weight and total activity calculation makes a LLW/TRU nCi/g determination for disposition purposes. Further development is being done to more efficiently automate the spreadsheet utilities.

Numerous legacy oversized waste items have been characterized for the 3706 Cubic Meter at LANL [8] using the Plan A approach. This approach has also been used in the newly generated decommissioning realm. The Plan B approach has not yet been utilized at LANL, largely because the limiting criteria for decontaminated transuranic waste, which is the majority of the waste so far characterized using the SCO techniques, is whether the waste meets low level criteria rather than whether it meets SCO criteria. The Plan C approach has been used extensively for legacy waste contaminated with depleted uranium.

PLAN A CALCULATIONS

The goal of Plan A is to estimate the maximum percent of potential survey points that may exceed the applicable SCO limit with an acceptable level of confidence. The collection of survey results from randomly selected locations ensures that the total data set will tend to be like the total surface area being evaluated with the degree of similarity between the sample and the represented population growing larger as the sample size increases. The choice of minimum sample size of 30 is based in using the sign-test (described in MARSSIM) [9]. The Plan A method evaluates several statistical parameters. The parameter used to calculate the total activity of the waste based on the samples is the 95% upper confidence level (UCL95). Table 2 shows the level of confidence for a sample size of n=30 and the proportion of population which may exceed the SCO limit of contamination given that no observation in the sample exceeds the SCO limit.

Table 2. Level of Confidence that Proportion is No Greater than P% for Sample of Size 30, Given that No Observation Exceeds SCO Limit

Maximum Percent (P%) Exceeding SCO Limit	Confidence Level (%)
1	26.0
2	45.5
3	59.9
5	78.5
7	88.7
10	95.8
15	99.2
20	99.9

This method requires that none of the 30+ samples or the UCL95 may exceed the applicable SCO limit. Also, neither the median nor the standard deviation may exceed ½ the SCO limit. These additional restrictions serve to strengthen the confidence that only a small proportion, if any, of the material being sampled exceeds the SCO limit - the effect of restrictions of known or expected distributional parameters is an approximately 95% confidence level that the proportion of contaminated surface area exceeding the SCO limit is less than 5%. The development of this conclusion is fully presented in references [6] and [7].

PLAN B CALCULATIONS

Plan B takes advantage of the observed data distributional features to generate more definitive quantitative characterization results. This is particularly helpful when the observed activity levels are near the SCO limits and the Plan A requirements of the median and standard deviation values not exceeding ½ the SCO are unlikely to be met. For Plan B, the characterization of the level of contamination is based on two separate but related criteria: (1) the greatest value the mean might be reasonably expected to have and (2) the greatest value some randomly selected sample point might reasonably be expected to display. The first criterion is applied by using the observed characteristics of the sample data to mathematically model the distribution of the largest value that the true mean (if the entire surface area were measured) would be expect to have. The second criterion is applied by calculating an upper tolerance limit (UTL) value for which there is a 90% confidence level that at least 98% of the surfaces in the sampled population will be less than the calculated value. The UTL is calculated using a method described in by Palachek [10].

The UTL can be calculated for either a normal or log-normal distribution and the method requires testing the data for normality or log-normality (here, D'Agostino Omnibus test). A description of the derivation of the application specific UTL calculation and the tests for normality (or log-normality) is fully presented in references [6] and [7].

DETERIMINING THE ACTIVITY OF LEGACY OVERSIZED TRU WASTE

We will concentrate on the legacy oversized items from plutonium operations since this is the bulk of the waste being decontaminated and characterized using SCO sampling. Figure 1 is an example of the glovebox waste as it was originally packaged in a fiber reinforced plywood box (FRP).

Figure 1. Fiber reinforced plywood boxes in storage containing legacy plutonium gloveboxes.

Figures 2 and 3 show the typical contents of an FRP as they might be seen as unpackaged in the decontamination facility at LANL. These photos were taken when the decommissioned gloveboxes were packaged for interim storage.

Figure 2. Photo of contents of a typical FRP.

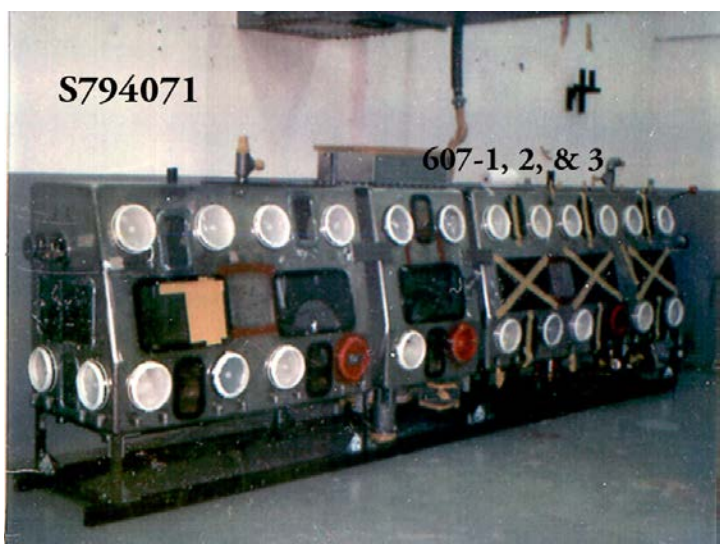

Figure 3.   Photo of legacy glovebox in above FRP.

Once the oversized waste is successfully unpackaged in the decontamination facility, a survey map is prepared and initial surveys are taken to determine the level of contamination in the waste. The same map is used after decontamination for the final survey of the waste for radioactive characterization. An example of a survey map for a decontaminated glovebox is shown below in Figure 4, with 33 survey points outside the box (accessible) and 33 survey points inside the box (inaccessible).

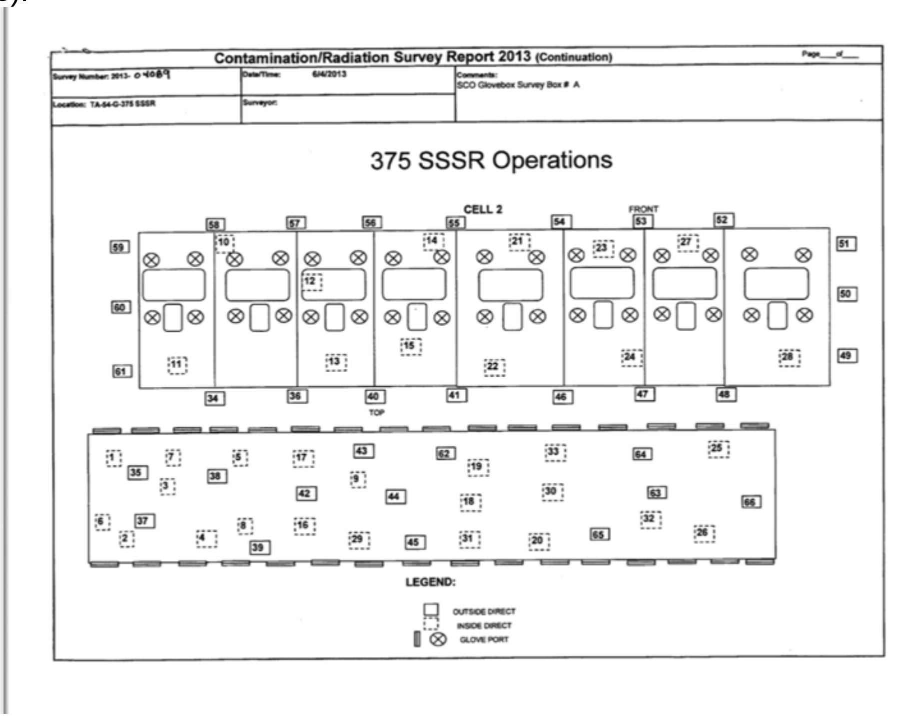

Figure 4. Example survey map for glovebox using Plan A.

Box dimensions and mass are provided to the analysis team by the decontamination crew and included with the box data package and survey results. For these items, calculations proceed using the UCL95 as the basis for determining the amount of activity in the material being characterized, while also meeting the limits for each data point, the median, and the standard deviation. The UCL95 (in disintegrations per minute per area) is multiplied by the applicable calculated surface area. Separate calculations are done for accessible surfaces (removable and fixed contamination) and the inaccessible surfaces (sum of removable and fixed contamination) to assure that SCO limits are met. The package total transuranic alpha activity is calculated based on the previously determined isotopic distribution. The mass of the waste item(s) is then used to determine if the transuranic concentration is less than 100 nCi/g and therefore allow the waste to be transported and disposed of as low level waste. To date, 1141 cubic meters of oversized legacy TRU waste has been decontaminated and prepared for disposal as low level waste using this method. Initial concentrations of slightly more than 100 nCi/g to 1000's of nCi/g have been decontaminated to less than 100 nCi/g, with an average decontamination factor of 50.

DETERIMINING THE ACTIVITY OF LEGACY OVERSIZED DEPLETED URANIUM WASTE

Another large waste stream which was characterized using the SCO process at LANL was contaminated with depleted uranium (largely U238). This waste stream included concrete blocks, metal sheets and contaminated equipment. The bulk of this waste stream consisted of concrete blocks that were characterized using Plan C – that is, the entire block was surveyed. The equipment characterized by sampling included several vehicles and a forklift. The level of uranium activity on all of this waste was very low and no decontamination efforts were required. The total volume of this low level waste stream characterized using SCO was approximately 175 cubic meters.

CONCLUSION

This SCO characterization process has been used successfully on a large volume of legacy TRU oversized waste items that have been decontaminated using one of several decontamination agents as described in reference [8]. The need to cut up or disassemble the oversized legacy waste to fit into acceptable WIPP containers has been greatly diminished, reducing the time, manpower, and risk associated with preparing the waste for transportation and disposal. The process has also been used successfully on a large volume of legacy depleted uranium waste that did not require decontamination. It is a very effective method of characterizing radioactive waste that is surface, rather than volume, contaminated. A few newly generated waste gloveboxes have been decontaminated in the plutonium facility and disposed of using SCO characterization – this is the expected route for process glovebox waste generated in the future.

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