RAPID/LOW-COST RADIOLOGICAL CHARACTERIZATION OF RESIDUAL URANIUM AND TECHNETIUM IN CONTAMINATED SCRAP METAL PILES[a]

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ABSTRACT

A large amount of contaminated scrap metal was generated during operations of the Oak Ridge Gaseous Diffusion Plant. Five scrap metal piles, consisting of approximately 40,000 tons of uranium-contaminated steel and aluminum, were rapidly characterized using *in situ* high-resolution gamma spectroscopy in combination with the Canberra *In Situ* Object Counting System (ISOCS) efficiency modeling software. ISOCS gamma spectroscopy was chosen in order to achieve direct measurement of large volumes of the piles (thus decreasing potential sampling error), accelerate completion of the project, minimize the safety risks of direct personnel contact with the scrap metal, and cut project costs by eliminating sample preparation and off-site shipping charges. A Canberra HPGe detector was attached to an 80’ boom and positioned over specific points on the surface of the piles. Forty-two separate measurements were performed, directly covering ~50% of the total surface area. For each point, a detection limit of ~1 pCi/g U-238 was achieved over a 30’-diameter circular field of view with a depth of 6-12”.

The results show that the scrap metal piles contain on average less than 2 pCi/g of U-235, less than 20 pCi/g of U-238, and very low concentrations of Co-60, Cs-137, Th-230, and a few other radionuclides [1]. These concentrations are well below the Waste Acceptance Criteria (WAC) limits for on-site disposal of 1500 pCi/g for U-235 and 1200 pCi/g for U-238 [2]. However, Tc-99 was also known to be a contaminant in this waste stream, and was in fact the limiting radionuclide for meeting the WAC requirements. Since Tc-99 does not emit gamma rays, an upper bound for the Tc-99 level was calculated by collecting 442 random metal pieces from the piles, scanning each piece with a hand-held beta probe, and making the conservative assumption that 100% of the counts were due to Tc-99. This upper bound was less than 10 pCi/g, well below the WAC limit of 172 pCi/g [2].

A study of cost- and time-savings compared to conventional sampling and off-site analysis showed that the combined *in situ* gamma spectroscopy and beta scanning strategy reduced characterization costs by 70% and the total project duration by 38%. This project therefore provides an example of the utility of ISOCS *in situ* gamma spectroscopy and field scanning measurements for accelerated and cost-effective waste disposition.

INTRODUCTION

The K770 scrap yard currently stores potentially surface contaminated scrap metal, the majority of which was generated during the operating life of the diffusion plant during the Cascade Improvement Program/Cascade Upgrade Program (CIP/CUP). At present it is estimated that approximately 40,000 tons of scrap metal are stored at K-770. In the 1980's, much of the scrap metal was segregated and size reduced. The metal was segregated into groupings of ferrous metals, non-ferrous metals, "special" metals with potential recycle value, and rubbish. Most of the scrap metal passed through the decontamination facility prior to storage. There, the scrap metal was vacuumed and decontaminated using water with dilute nitric acid or an alkaline detergent. The standard of this decontamination effort was to remove transferable uranium prior to outside storage. The primary known radiological contaminants of concern are low-enriched uranium and Tc-99.

An aerial photo of the five scrap piles 14A, 14B, 16A, 16B, and 16C, addressed in this project is shown in Fig. 1. Pile 14A consists of shredded Al and 14B is non-shredded Al disks. The 16A pile is made up of Ni-plated iron from the Oak Ridge Gaseous Diffusion Plant. The 16B and 16C piles are made up of contaminated iron from the Y-12 plant.

Under the current DOE Oak Ridge accelerated cleanup initiatives, the metal is scheduled for accelerated disposition in 2004, preferably to the Environmental Management Waste Management Facility (EMWMF) in Oak Ridge. DOE is also emphasizing innovative characterization approaches that significantly reduce costs and increase characterization flexibility without degrading data quality.

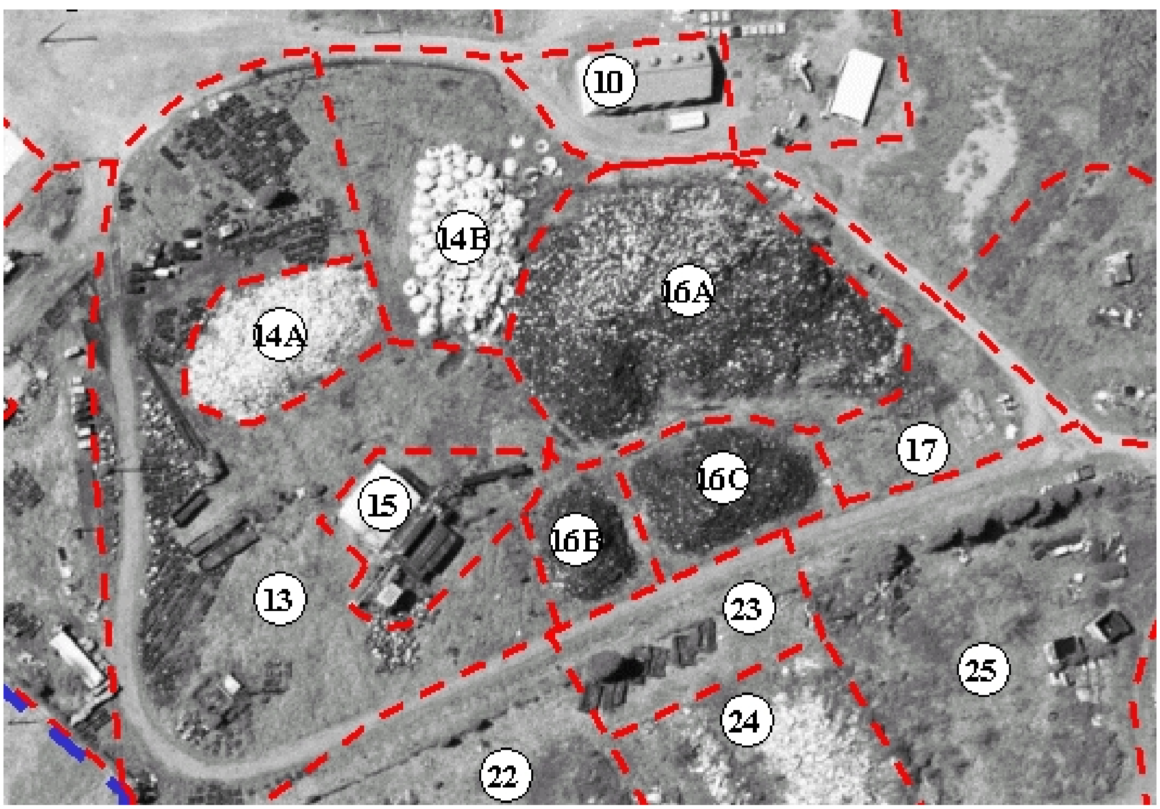

Fig. 1 Aerial view of the five large metal scrap piles (14A, 14B, 16A, 16B, and 16C) located in the K-770 scrap yard in Oak Ridge, TN.

NDA APPROACH FOR URANIUM QUANTIFICATION

An ISOCS high-resolution gamma spectroscopy system was used for the in-situ NDA measurements. The ISOCS system was chosen for its portability, high sensitivity, and ability to measure and analyze a wide range of sample geometries. ISOCS utilizes a mathematical calibration approach that eliminates the need for calibration sources or sample surrogates [4]. ISOCS has been used to date to support a wide variety of environmental restoration and decommissioning/decontamination projects [5,6]. Canberra Oak Ridge routinely uses ISOCS systems to characterize radioactive constituents in waste containers, in piping and process equipment, in soil, and in decommissioned UF_6 cylinders [7].

The detector was a coaxial germanium detector with a 180-degree field of view 50mm-thick lead collimator. The 50mm of lead side-shielding reduced interferences from adjacent scrap piles, contaminated soil, and other contaminated items in the area. The 180-degree collimator allowed for a wide field of view for each measurement of the scrap piles. The width of the field of view depends strongly on the distance of the detector from the surface of the pile. A larger detector offset yields a wider field of view; however, the detection sensitivity decreases. With a 5 foot offset from the surface of the pile, the measured surface area was approximately 30 feet in diameter. This offset was chosen as the best balance between sensitivity and field of view for this application. Low MDA's were achieved (see Table I) while still keeping the number of measurements required to characterize the total surface of a pile to a manageable number.

A photograph of the ISOCS detector, securely mounted in the boom basket, is shown in Fig. 2. The shiny cylinder on the left is the lead shielding, with the germanium detector mounted concentrically inside of it. The beige cylinder on the right is a small liquid nitrogen Dewar to keep the detector cold. The system could be tilted or rotated to nearly any orientation. Data collected by the spectroscopy system were transferred to a PC on the ground via a wireless modem. Six flexible poles were mounted on the front of the boom basket so that the detector could be positioned a constant 5 feet from the face of the piles with an orientation approximately normal to the surface of the piles.

Each of the scrap piles was divided into wedge-shaped sections. The largest 16A steel pile had 10 sections, the intermediate 14A, 14B, and 16C piles had six sections each, and the smallest 16B pile had four sections. Two ISOCS measurements were performed in each section of the 16A pile, one near the ground and the second near the top of the pile, for a total of 20 measurements. One ISOCS measurement was performed in each of the other 22 sections of the other four scrap piles. Figure 3 shows a photograph of the ISOCS detector deployed on the end of the boom for a measurement on the large 16A scrap pile. Figure 4 shows a close-up of one of the 16A pile measurements.

Fig. 2 ISOCS detector mounted in the basket of the 80-foot boom

Fig. 3 Photograph of ISOCS detector deployed in boom basket over the 16a steel pile

Nondestructive Assay Results

Average Minimum Detectable Activities (MDA’s) for the ISOCS measurements, as calculated by the Canberra Genie2k gamma spectroscopy analysis software, are shown in Table I. Very low detection limits were achieved throughout the project through the use of relatively long counting times and 50mm-thick lead shielding around the detector to reduce gamma interferences.

Table I Calculated Average MDA’s for the ISOCS Measurements of the Five Scrap Piles

Pile	**U-235 MDA pCi/g**	**U-238 MDA pCi/g**	**Co-60 MDA pCi/g**	**Cs-137 MDA pCi/g**	**Th-230 MDA pCi/g**	**Th-232 MDA pCi/g**	**Np-237 MDA pCi/g**
16A	3.59E-02	9.82E-01	3.90E-03	9.53E-03	2.22E-02	1.52E-02	3.26E-02
16B	3.90E-02	1.18E+00	1.53E-03	9.90E-03	2.74E-02	2.00E-02	3.74E-02
16C	6.73E-02	1.21E+00	5.13E-03	1.26E-02	3.06E-02	1.82E-02	4.09E-02
14A	4.85E-02	1.48E+00	5.39E-03	2.49E-02	3.42E-02	2.01E-02	5.80E-02
14B	8.46E-02	1.57E+00	5.98E-03	1.92E-02	3.44E-02	1.89E-02	5.11E-02

A summary of the NDA results is given in Table II for the five scrap metal piles. In addition to U-238 and U-235, other radionuclides, including Cs-137, Co-60, Np-237, Th-230, and Th-232, were detected and quantified in some of the measurements. The table also shows the relevant EMWMF WAC criteria. The Analytic WAC are derived from the approved risk assessment model in the remedial investigation/feasibility study [8]. The Analytic HI WAC are based on the risk assessment models for the Hazard Index of non-carcinogenic constituents. The Analytic Carcinogenic WAC are based on the risk assessment models for carcinogenic constituents. The ASA WAC are derived from the facility authorization basis documentation for the EMWMF. For these radionuclides, the most limiting WAC is the Analytic Carcinogenic WAC of 1200 pCi/g for U-238.

Several conclusions can be drawn from these results:

- U-238 and U-235 were the dominant gamma-emitting radioactive contaminants in this waste stream, as expected
- The uranium was in the form of low-enriched uranium (~1%) for all five scrap piles and the measured enrichments were very consistent for the five piles
- Cs-137, Co-60, Np-237, Th-230, and Th-232 were only present at very low “trace” concentrations
- All measured concentrations were well below the relevant WACS. The lowest WAC:measured ratio was 37:1 for U-238 in the 14A scrap pile.

A contour map of the measured U-238 concentrations on the surface of the 16A steel scrap pile is shown in Fig. 4. The U-235 measurements show a very similar distribution. The contours indicate that the uranium concentrations vary slowly and smoothly over the surface of the pile, with no large hotspots of contamination. This supports the hypothesis that the uranium distribution in the pile is relatively homogeneous.

Table II NDA results (in Pci/G) for eight radionuclides measured in the five scrap metal piles. The relevant EMWMF WAC are also shown.

Scrap Pile	Radionuclide	Mean	Standard Deviation	Analytic HI WAC	Analytic Carcinogenic WAC	ASA WAC
14A	Cs-137	0.0174	0.018	NA	NA	1.5E6
	Co-60	0.0021	0.0015	NA	NA	6.6E6
	Np-237	0.109	0.0504	NA	NA	10,000
	Th-230	0.068	0.0315	NA	NA	15,000
	Th-232	0.106	0.0797	NA	NA	2500
	U-235	1.92	0.687	9500	1500	100,000
	U-238	32.2	7.13	1500	1200	100,000
	U enrichment %	0.889	0.209	NA	NA	NA
14B	Cs-137	0.016	0.0095	NA	NA	1.5E6
	Co-60	9.2E-4	6.1E-4	NA	NA	6.6E6
	Np-237	0.17	0.0554	NA	NA	10,000
	Th-230	0.093	0.0279	NA	NA	15,000
	Th-232	0.114	0.0551	NA	NA	2500
	U-235	0.649	0.243	9500	1500	100,000
	U-238	11.1	3.19	1500	1200	100,000
	U enrichment %	0.892	0.181	NA	NA	NA
16A	Cs-137	0.0109	0.0073	NA	NA	1.5E6
	Co-60	0.0038	0.0047	NA	NA	6.6E6
	Np-237	0.0384	0.0413	NA	NA	10,000
	Th-230	0.0474	0.0259	NA	NA	15,000
	Th-232	0.0727	0.0632	NA	NA	2500
	U-235	1.09	0.606	9500	1500	100,000
	U-238	17.1	9.0	1500	1200	100,000
	U enrichment %	1.03	0.308	NA	NA	NA
16B	Cs-137	0.0659	0.0308	NA	NA	1.5E6
	Co-60	0.0015	6.6E-4	NA	NA	6.6E6
	Np-237	0.0313	0.0214	NA	NA	10,000
	Th-230	0.114	0.0553	NA	NA	15,000
	Th-232	0.205	0.123	NA	NA	2500
	U-235	0.87	0.249	9500	1500	100,000
	U-238	12.9	4.62	1500	1200	100,000
	U enrichment %	1.10	0.449	NA	NA	NA
16C	Cs-137	0.0972	0.0522	NA	NA	1.5E6
	Co-60	0.0018	9.2E-4	NA	NA	6.6E6
	Np-237	0.054	0.0249	NA	NA	10,000
	Th-230	0.0995	0.0376	NA	NA	15,000
	Th-232	0.315	0.136	NA	NA	2500
	U-235	1.10	0.911	9500	1500	100,000
	U-238	15.3	3.47	1500	1200	100,000
	U enrichment %	1.12	0.966	NA	NA	NA

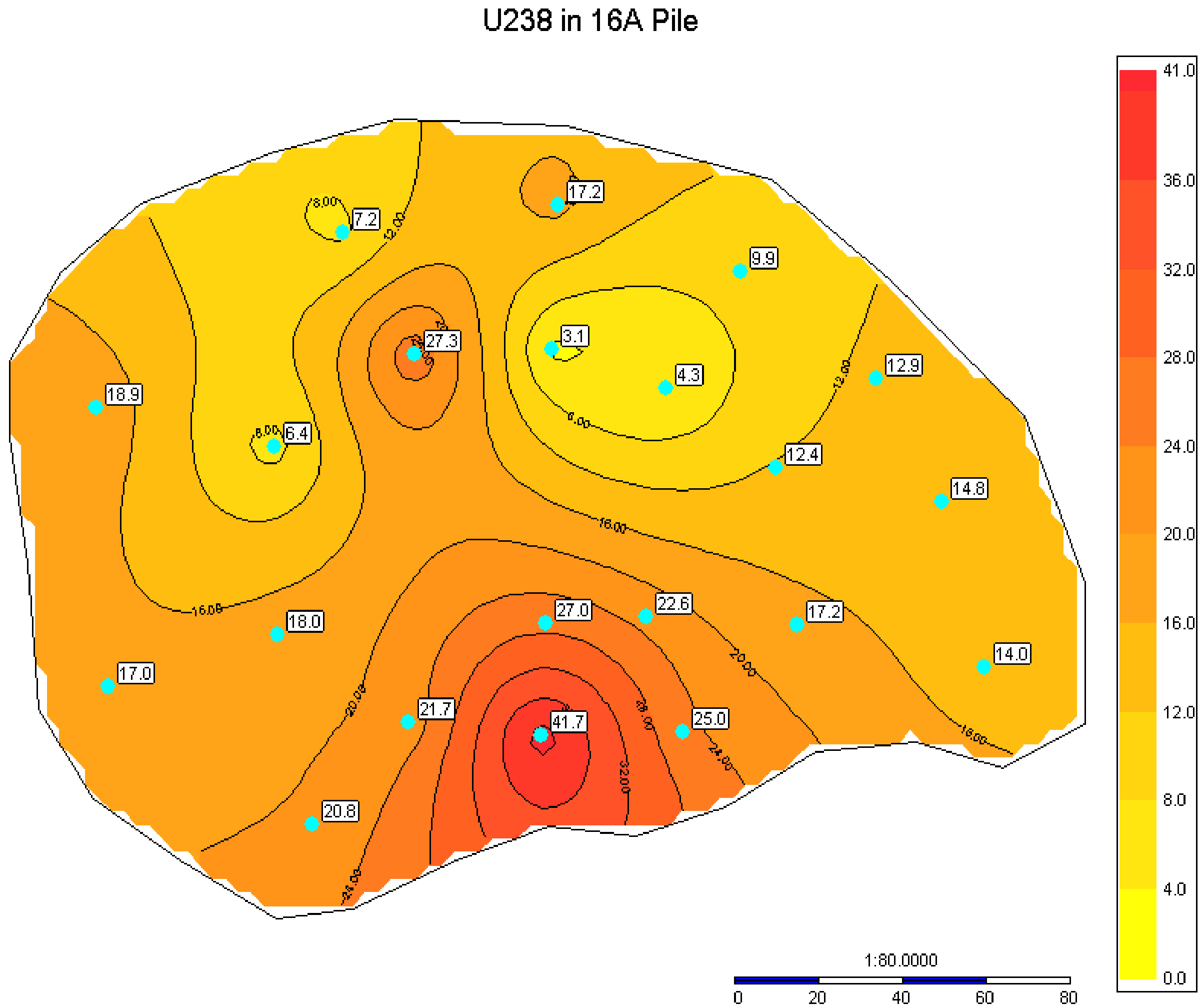

Fig. Contour map of measured U-238 concentrations on the surface of the 16A steel sScrap pile. Units are Pci/G. The blue dots indicate measurement locations and values.

BETA SCANNING APPROACH TO QUANTIFY TC-99 CONCENTRATIONS

In parallel with the NDA measurements, Canberra collected 413 individual items off of the 16A, 16B, 16C, and 14A scrap piles for beta scanning. Every effort was made to make the collection as random as possible, within the following constraints:

- items had to be light enough for one person to lift and move without strain
- items had to be small enough to handle manually without becoming a safety hazard

- items had to have a sufficiently regular shape to allow for easy measurement of external dimensions
- items had to have surfaces which could be 100% accessible by the handheld beta probe (e.g. no small-diameter pipes)
- items were lying on the surface, not partially or fully buried
- items had to have sufficient integrity to allow for handling and movement to the staging area

After collection the items were moved to a covered staging area near the 16A pile. Prior to beta scanning, each item was measured and its external dimensions were recorded on logsheets. After beta scanning, the items from each section were grouped together and stacked on pallets. Plastic sheeting was placed between layers on the pallets to prevent cross-contamination.

For the large aluminum disks in the 14B pile, the items were too large, heavy, and bulky to be moved by hand, so the beta scanning measurements were performed with the items in place on the pile. Ten different disks were scanned.

Beta Scanning Measurement Approach

Beta scanning measurements were performed with a Ludlum model 44-9 beta probe attached to a model 2221 scaler. The beta probe was calibrated for Tc-99 using a traceable planar Tc-99 source.

The following steps were performed for each item:

1) The item was scanned for hotspots of beta activity.
2) If one or more hotspots were found, then the hotspot with the highest beta count rate was measured for 30 seconds. The position of the reading was marked on the item and the gross beta count was recorded on the beta scanning logsheet. Up to nine other locations were selected at random, marked, measured for 30 seconds, and recorded. If the item was not large enough to allow for ten separate measurements, then as many separate beta measurements were performed as the size of the item allowed.
3) If no hotspots were found, then up to ten locations were selected at random, marked, measured for 30 seconds, and recorded. If the item was not large enough to allow for ten separate measurements, then as many separate beta measurements were performed as the size of the item allowed.

For the 14B disks, the items were measured in place on the pile. The same protocol was followed for each item with ten measurements performed on the inside surface and ten measurements on the outside surface of each item.

Estimated Upper Bound Tc-99 Concentrations

The calculation of the total Tc-99 specific activity for each metal item was performed as follows:

1) The total surface area was calculated from its measured external dimensions.

2) The total mass of the item was calculated based on calculated volume and know bulk density.
3) The average net (gross – background) beta count rate was calculated for the item.
4) It was assumed that 100% of the measured net beta counts were due only to Tc-99. It is possible that uranium or thorium daughters contributed to the measured beta count rates. This assumption yields a **conservative upper limit** for the estimated Tc-99 concentrations. The average net beta count rate for the item was then converted to average Tc-99 activity per sq. cm. using the known detector efficiency for Tc-99.
5) The total Tc-99 activity for the item was then calculated by multiplying by the total surface area of the item.
6) The total Tc-99 specific activity for the item was then calculated by dividing by the estimated mass of the item.

The upper bound estimated average Tc-99 concentrations for each scrap pile are listed in Table III. MDA's were calculated with Curie formula [9] using background count rates on a clean piece of steel and the average size and mass of items from the particular piles. The relevant WAC's are also shown. The limiting WAC is the Analytic Carcinogenic WAC of 172 pCi/g. The aluminum 14A and 14B scrap piles exhibited Tc-99 concentrations that were more than ten times higher on average than the 16A, 16B, and 16C steel scrap piles. The average Tc-99 concentrations for two of the three steel piles were below the estimated MDA's for those piles. The aluminum 14A pile had the highest average Tc-99 specific activity of 47.7 pCi/g and a comparatively large standard deviation of the measurements of 110 pCi/g (231% relative standard deviation). Seven of the individual items measured from the 14A pile exceeded the 172 pCi/g limiting WAC value. None of the other individual 327 items from the other four piles exceeded this value. The highest measured Tc-99 specific activity for any item was 776 pCi/g for one item from the 14A scrap pile.

Table III Beta scanning results (in Pci/G) for Tc-99 in the five scrap piles. The relevant EMWMF WAC are also shown. The number of individual measurements that exceeded the limiting WAC is also shown.

Scrap Pile	Mean	Standard Deviation	Average MDA	Analytic HI WAC	Analytic Carcinogenic WAC	Frequency> Carcinogenic WAC	ASA WAC
14A	47.7	110	3.89	NA	172	**7/96**	4.2E7
14B	42.1	26.0	5.68	NA	172	0/10	4.2E7
16A	1.24	11.8	2.50	NA	172	0/158	4.2E7
16B	2.30	2.82	2.50	NA	172	0/64	4.2E7
16C	3.60	6.39	2.50	NA	172	0/95	4.2E7

ANALYSIS OF COST AND TIME SAVINGS

Traditional Sampling and Analysis Methodology

The baseline characterization scenario for the purpose of cost and duration comparisons assumes that the five metal scrap piles are sampled using traditional sampling methods, and that the samples are transported offsite to a radiochemical laboratory for analysis. The approach would be described in detail in a Sampling and Analysis Plan. Using the 80-foot boom, 56 steel and aluminum items would be collected at random from the surfaces of the 14A, 16A, 16B, and 16C scrap piles. 14 coupons would be cut at random from the large aluminum disks in the 14B scrap pile and analyzed at the offsite laboratory.

A summary of the duration and costs of the major subtasks in this baseline scenario is given in Table IV. The turnaround time for the offsite laboratory analysis of any individual item is 14 working days. The dominant cost factor in this scenario is the cost for offsite radiochemical analyses. The total baseline project duration is estimated to be 52 working days and the total estimated cost is $237,800.

Table IV Estimated duration and costs for the baseline sampling and laboratory analysis characterization scenario

Task Description	Time (working days)	Cost
Develop Sampling & Analysis Plan	10 days	$15,000
Mobilization/demobilization	2 days	$800
Collect steel & aluminum samples	5 days	$4,500
Cut coupons from Al disks	2 days	$14,000
Sample transport	3 days	$8,000
Laboratory radiological analyses	30 days	$184,500
Statistical analyses	14 days	$11,000
total	52 days	$237,800

Nondestructive Assay and Beta Scanning Costs and Duration

A summary of the duration and costs of the major subtasks in the NDA/beta scanning characterization scenario, as described in this paper, is given in Table V. The turnaround time for an individual NDA measurement analysis is 0.5 – 1 day. The dominant cost contributors are the cost for performing the NDA measurements, the cost for performing the beta scanning measurements, and the cost for analyzing the NDA measurements. The total NDA/beta scanning project duration was 32 working days and the total cost was $70,300.

Table V. Actual Duration and Costs for the NDA/Beta Scanning Characterization Scenario.

Task Description	Time (working days)	Cost
Develop NDA Plan	5	$4,200
Mobilization/demobilization	2	$800
Perform NDA measurements	11 days	$18,400
Collect steel & aluminum samples	5 days	$4,500
Beta scanning of samples samples	10 days	$16,700
NDA analyses & report	26 days	$21,700
Statistical analyses	5 days	$4,000
total	32 days	$70,300

Advantages of the NDA/Beta Scanning Methodology

The advantages of the NDA/beta scanning approach, compared with the traditional sampling and analysis approach, include:

1) Analysis turnaround times are typically less than one day, compared with 14 days, allowing for a flexible and responsive characterization approach.
2) 70% reduction in characterization costs
3) 38% reduction in total project duration
4) The NDA measurements are more representative than the sampling measurements. The volume of metal “sampled” in a single ISOCS measurement is much greater than the volume sampled from any single metal item collected from the scrap piles. The typical NDA ‘sample volume’ in this measurement geometry is on the order of 800 cubic feet (2.2E7 cm^3). A typical laboratory sample volume is on the order of 100 cm^3. In a non-homogeneous waste stream such as scrap metal, many more samples would need to be collected than NDA measurements to achieve the same degree of representativeness.

CONCLUSION

Canberra Oak Ridge developed and applied an innovative measurement methodology, combining in-situ NDA measurements and beta scanning, to the radiological characterization of five large scrap metal piles located at the East Tennessee Technology Park in Oak Ridge, TN. Forty-three high-resolution gamma spectroscopy measurements were performed on the five piles. These ISOCS measurements quantified the concentrations on U-238, U-235, and very low levels of Cs-137, Co-60, Np-237, Th-230, and Th-232 in those piles. Beta scanning was performed of 422 metal items from the five piles. Analysis of these measurements yielded conservative upper bounds for the concentrations of Tc-99 in the five piles. These analyses indicate that the concentrations of radioactive constituents in the 16A, 16B, and 16C steel piles are far below the waste acceptance criteria for the EMWMF disposal facility in Oak Ridge and are likely to be good candidates for disposition to that facility. The beta scanning measurements indicate that the Tc-99 concentrations in the 14A and 14B aluminum scrap piles are a significant fraction of the EMWMF WAC limit and may not be suitable candidates for disposal to that facility. The characterization project was completed in 32 working days at a cost of $70,300. This is a 70%

reduction in cost and a 38% reduction in duration over a baseline sampling and laboratory analysis approach.

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FOOTNOTES

[a] This work was performed by Canberra Oak Ridge, LLC for the United States Department of Energy under contract to Bechtel Jacobs Company, contract 23900-BA-LW023U.