Lessons Learned from the Application of Bulk Characterization to Individual Containers on the Brookhaven Graphite Research Reactor Decommissioning Project at Brookhaven National Laboratory - 12056

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ABSTRACT

When conducting environmental cleanup or decommissioning projects, characterization of the material to be removed is often performed when the material is in-situ. The actual demolition or excavation and removal of the material can result in individual containers that vary significantly from the original bulk characterization profile. This variance, if not detected, can result in individual containers exceeding Department of Transportation regulations or waste disposal site acceptance criteria.

Bulk waste characterization processes were performed to initially characterize the Brookhaven Graphite Research Reactor (BGRR) graphite pile and this information was utilized to characterize all of the containers of graphite. When the last waste container was generated containing graphite dust from the bottom of the pile, but no solid graphite blocks, the material contents were significantly different in composition from the bulk waste characterization. This error resulted in exceedance of the disposal site waste acceptance criteria. Brookhaven Science Associates initiated an in-depth investigation to identify the root causes of this failure and to develop appropriate corrective actions. The lessons learned at BNL have applicability to other cleanup and demolition projects which characterize their wastes in bulk or in-situ and then extend that characterization to individual containers.

HISTORY

Construction of the BGRR was completed in 1950 making it the first nuclear reactor built for the sole purpose of providing neutrons for research. The BGRR was an air-cooled graphite moderated reactor. During reactor operations, outside cooling air was drawn through the reactor pile, cooled, filtered and exhausted out of a 100-meter tall stack.

Originally the BGRR was fueled with natural uranium fuel slugs from 1950 to 1957/58. During this time there were 28 reported ruptured fuel slug and one experimental sample rupture. In 1957/58 the natural uranium metal fuel slugs were replaced with fuel cartridges which contained an alloy of aluminum and enriched uranium (93% U-235) and were clad in aluminum. The new fuel allowed for higher neutron flux and did not result in any reported fuel failures, although there was some minor loss of integrity resulting in surface contamination with tramp uranium and small routine releases of fission products.

The BGRR was retired from service in 1968 and all fuel was removed by 1972. Decommissioning of the graphite pile was initiated in December 2009 and removal of the graphite blocks complete in March 2010.

RADIOLOGICAL CHARACTERIZATION

In December of 2005 Brookhaven Science Associates issued the "Brookhaven Graphite Research Reactor Determination of Radionuclide Inventory of the Graphite Pile for Waste Stream Characterization and Waste Acceptance Criteria Compliance". The purpose of this report was to document the methodology, sampling, analysis and conclusions from previous graphite characterization work performed in 2000 and updated with supplemental sampling performed in 2005. Over the years since shutdown there had been several estimates of the amount of radioactivity contained in the BGRR graphite pile.

In 1998, the initial pile inventory estimate was 5.55E+04 gigaBequerels (GBq) and about 480% of the Nuclear Hazard Category 3 fraction as defined in DOE Standard 1027-92. This was a rough bounding estimate based on extrapolation of a study done in the United Kingdom which focused on planning for the decommissioning of graphite power reactors in Great Brittan.

In preparation for isolation of the pile in 2000, a more formal radiological characterization was performed to bound the risk associated with working in or around the graphite pile. A Technical Work Document provided a logical and planned approach to obtain pile characterization data needed to plan for the waste disposal and to assess potential health and safety hazards during stabilization and later decommissioning work. The plan called for a combination of radiological survey readings, In-Situ Object Counting (ISOCS), and analysis of graphite samples obtained from each of the five different graphite regions. This effort conservatively estimated the inventory of the pile to be less than 1.70E+05 GBq; which is greater than in 1998, however, the pile was now estimated to be only 350% of the Nuclear Hazard Category 3 fraction.

In 2005 the BGRR Record of Decision was signed by the United States Department of Energy, the United States Environmental Protection Agency and the New York State Department of Environmental Conservation. At this time the pile inventory was estimated at 1.20E+05 GBq. This agreement required removal of the BGRR graphite pile, therefore additional data was required for accurate characterization of the graphite pile as a waste stream and to eliminate some of the over-conservative bias inherent in the previous estimates which were aimed at bounding the radiological inventory. Laboratory analysis of 17 of the previously obtained smear samples taken from a combination of failed fuel channels, fuel channels and non-fuel channels along with analysis of 25 graphite samples that were obtained based on reactor design, location of the different grades of graphite, neutron flux symmetries, operating history and location accessibility were performed. It was now estimated that the BGRR graphite pile contained a total activity (as of 1-1-2007) of 2.91E+04 GBq in a volume of 442.5 cubic meters, for an average concentration of 65.76 GBq/m3, or 4.40E-05 GBq/g. The dominant radionuclide was determined to be C-14 (2.11 GBq). Other significant radionuclides identified included Ni-63, H-3, and Eu-152. Dispersible contamination in the pile included Sr-90, Cs-137, Am-241, and Pu-238, -239, -240 and -241.

In preparation for removal and packaging of the graphite blocks in 2010 a report titled the Updated Waste Characterization Procedure for Graphite Pile Waste Containers – December 2009 was issued to update the inventory and provide a methodology for determining activity in the graphite boxes was prepared. The activities calculated in the 2005 report were decay corrected leaving the estimated total curie content at 2.83 GBq, due to decreases in all of the relatively short-lived radionuclides. The C-14 activity did not substantively decrease by decay. The report prescribed an overall methodology for characterizing individual boxes of graphite that entailed three types of scaling calculations described below and detailed in Table-1.

For C-14 and H-3, activity was calculated by concentration and weight. Determination of C-14 was based on weight. The concentration of C-14 in the pile did not vary substantially in different locations. For example, the C-14, the concentration of type A graphite (closest to pile center) is less than two times the average, and the concentration of type D graphite (furthest from pile center) is about 58% of the average. Therefore, since the variability is not great, the average concentration of 3.19E-05 gBq/g was used for all the graphite.

H-3 has a similar low variability throughout the pile, where the type B graphite has the highest tritium concentration (2.4 times the average), and the type D graphite has the lowest concentration (30% of the average). Therefore, the average H-3 concentration of 3.68E+03 Bq/g was used for all the graphite. Each container of graphite was weighed and the amount of C-14 and H-3 was calculated using the above average concentrations.

For all other radionuclides except C-14, H-3, and the fissile radionuclides (U-235, Pu-239, and Pu-241), the measured dose rate at 1 meter from the container was used to calculate the activity in the container. MicroShield™ Version 7 was used to perform the calculations, using the radionuclide mix in the graphite as a method for scaling the activity. The gamma emitters were used in the MicroShield™ calculation, and other radionuclides are scaled to the gamma emitters. This scaling of radionuclides was a reasonable assumption because all of the radionuclides identified in the Updated Waste Characterization Procedure for Graphite Pile Waste Containers – December 2009 (except C-14 and H-3) are expected to be associated with the gamma emitters in failed fuel (e.g., Cs-137, Sr-90, alpha emitters) and in activated materials (e.g,Co-60, Ni-63).

The method for determining activity in a graphite box was gamma calculation. The dose rate from one graphite container was determined by using MicroShield with conservative assumptions of the weight of graphite, size of the box, density of graphite and graphite fill line. Although the graphite contains a mixture of radionuclides, only the gamma emitters were used in the calculations.

For the fissile radionuclides, U-235, Pu-239, and Pu-241, activity was calculated by weight, with a conservative estimation of activity. The characterization data was reviewed for the five different grades of graphite used, to determine the type of graphite that yielded the highest Fissile Grams Equivalent (FGE). For each isotope the highest concentrations were used to develop the conservative scaling factors.

In addition, the calculation was performed assuming that the highest concentrations in any type of graphite occurred together in each box. That is, that each box contained U-235 at the highest concentration of type AA graphite, Pu-239 at the concentration of type B graphite, and Pu-241 at the concentration of type A.

When using these concentrations, it was confirmed that no single box exceeded the 15 grams of fissile material requirement, and all shipments were determined to be exempt from the fissile packaging and transportation requirements.

The methodology describe above was used to calculate the Curie content of the graphite dust box as had been done for all prior graphite boxes. However, the documentation representing the dust box mis-stated the volume of waste. The box was described by the generator as being completely full. This incorrect volume caused errors in the calculations that were performed to verify compliance with the EnergySolution's Waste Acceptance Criteria for Class A waste limits.

Table 1 – Scaling Factors and Fractions of Total Activity in Graphite Box

Method of ID or Calculation	Radionuclide	GBq in Pile	Fraction of Total	Corrected Activity in Box (GBq)	Scaling Factors	Units
A. Calculated by weight	H-3	2.44E+03	8.59E-02	1.25E+01	3.68E-06	GBq/g
	C-14	2.11E+04	7.45E-01	1.08E+02	3.19E-05	GBq/g
B. Identified in Characterization Report	Co-60	1.24E+02	4.37E-03	6.36E-01	6.44E+00	GBq/mSv/hr @ 1m
	Ni-59	1.86E+05	6.57E-06	9.55E-04	9.66E-03	GBq/mSv/hr @ 1m
	Ni-63	3.74E+03	1.32E-01	1.92E+01	1.94E+02	GBq/mSv/hr @ 1m
	Sr-90	3.48E+01	1.23E-03	1.78E-01	1.81E+00	GBq/mSv/hr @ 1m
	Tc-99	1.85E-01	6.54E-06	9.51E-04	9.62E-03	GBq/mSv/hr @ 1m
	I-129	1.91E-01	6.72E-06	9.77E-04	9.88E-03	GBq/mSv/hr @ 1m
	Ba-133	1.21E+01	4.28E-04	6.22E-02	6.29E-01	GBq/mSv/hr @ 1m
	Cs-137	2.48E+01	8.75E-04	1.27E-01	1.29E+00	GBq/mSv/hr @ 1m
	Eu-152	6.55E+02	2.31E-02	3.36E+00	3.40E+01	GBq/mSv/hr @ 1m
	Eu-154	1.58E+02	5.59E-03	8.10E-01	8.21E+00	GBq/mSv/hr @ 1m
	Eu-155	8.07E+00	2.85E-04	4.14E-02	4.18E-01	GBq/mSv/hr @ 1m
	Ra-226	2.55E-01	8.98E-06	1.31E-03	1.32E-02	GBq/mSv/hr @ 1m
	Th-232	7.66E-02	2.70E-06	3.92E-04	3.96E-03	GBq/mSv/hr @ 1m
	U-234	2.65E-01	9.35E-06	1.36E-03	1.38E-02	GBq/mSv/hr @ 1m
Fissile – by weight	U-235	2.56E-02	9.05E-07	3.06E-04	8.97E-11	GBq/g
	U-238	2.25E-02	7.92E-07	1.15E-04	1.17E-03	GBq/mSv/hr @ 1m
	Pu-238	1.91E+00	6.74E-05	9.81E-03	9.92E-02	GBq/mSv/hr @ 1m
Fissile – by weight	Pu-239	3.04E+00	1.07E-04	9.32E-02	2.73E-08	GBq/g
Fissile – by weight	Pu-241	1.57E+01	5.52E-04	1.81E-01	5.31E-08	GBq/g
	Am-241	3.92E+00	1.38E-04	2.01E-02	2.03E-01	GBq/mSv/hr @ 1m
	Total	2.83E+04	1.00E+00	1.46E+02		

DISCUSSION

Approximately two hundred and fifty (250) boxes of graphite debris were generated and characterized for disposal using the methods specified above. All of the boxes of solid graphite blocks and debris were shipped to disposal without incident.

In May 2010 generation of a box of waste with material from the vacuuming of the bottom of the graphite pile was initiated. The box then received more waste from a second vacuuming event performed in December 2010 after the Air Tight Membranes and Neutron Shields were removed from the pile area. Because the vacuuming tended to deform the walls of the box, a metal frame was inserted by ERP to support them laterally. The box was finally closed in June of 2011 and shipped to EnergySolutions. This container was characterized using the same methodology approved for the graphite blocks, as if it consisted of graphite blocks from the pile.

This last container of waste that was removed from the graphite pile had mistakenly been assumed to have the same isotopic content and distribution consistent with the graphite pile as a whole. The physical methodology used to remove the graphite blocks from the pile knocked the blocks around and hence cause the mobile contamination in the form of dust that had been coating the blocks to fall off and drift to the bottom of the pile. Not only did this dust concentrate the contamination, but it was much lighter and did not have the same density of the solid graphite blocks. The high degree of contamination that had been present on the surface of the graphite blocks due to fuel failures in the past increased the fission product content of the dust that accumulated at the bottom of the pile.

Upon arrival at EnergySolutions of Utah, the box was sampled and was found to exceed their Waste Acceptance Criteria. Specifically, the waste was greater than Class A.

A root cause Investigation Committee was appointed to investigate the event, determine root cause(s) and develop corrective actions to minimize the risk of recurrence. The Investigation Committee included representatives from several technical Divisions and the Quality Management Office. The Committee also included a trained TapRooT Advanced Investigation Team Leader. The Committee was chaired by a Certified Health Physicist. The DOE was informed of the Committee's activities and observed the meetings.

The Investigation Committee employed the TapRooT methodology, the Barrier Analysis methodology, and an Events and Causal Factors Chart to determine causes of this event, along with previous associated events relevant to radioactive waste characterization at BNL. The methods used focused on the system, not on the actions of any individual.

Five causal factors were identified using the Events and Causal Factors chart, and then individually evaluated using the TapRooT methodology, which resulted in the identification of associated TapRooT causes. The TapRooT causes were mapped to the Occurrence Reporting Processing System (ORPS) cause codes. The Barrier Analysis Method was used to confirm that corrective actions determined through the previous two methods addressed the failed barriers identified by the Barrier Analysis.

- Causal factor (CF) #1 – The weight and volume was reported incorrectly on characterization paperwork

- CF #2 - Generator did not recognize waste item as different from previous boxes of graphite blocks and therefore used inappropriate characterization and did not recognize need to sample box contents.
- CF #3 - Extent of condition review from previous incident (Ref. ORPS SC--BHSO-BNL-BNL-2010-0016) inadequate because it did not require a review of waste already in storage
- CF #4 - Generator did not sample the waste as required by recently revised procedure
- CF #5 - Waste Class calculations were not redone when the volume of waste was corrected on the manifest after verbal information was received by the shipper just prior to shipment.

The team also performed a Barrier analysis of the event, with a trained facilitator. The Barrier analysis identified a number of barriers which had failed:

- The waste generator and waste management reviewer understanding of characterization basis and radiochemistry was less than adequate
- Oversight/second check of the data review was lacking
- Procedures/training less than adequate including the site-wide standards based management system, the waste management and the waste generator procedures
- Waste management reviewers were inconsistent in their understanding of their roles with regard to data/paperwork reviews

The Investigation Team evaluated all of the analyses and identified a substantial number of corrective actions to address the root and apparent causes and failed barriers. Corrective actions which may have general applicability to other sites and projects include:

- Assure that waste management is staffed with qualified and experienced radioactive waste characterization analysts.
- Establish agreement with another DOE site for peer review of select waste stream characterization (i.e. >50% Class A limits, >2 mSv/hr, >100,000 dpm alpha).
- Institute review of radioactive waste shipments by a committee including expertise in Health Physics, Transportation, Radiological Controls and the targeted disposal facility's Waste Acceptance Criteria.
- Require that smears of internal contents of containers be taken and counted for alpha and beta/gamma, that the count results be compared to the stated characterization.
- Require confirmatory gamma spectroscopy be performed for packages exhibiting dose rates of greater than 0.5 mSv/hr on contact.

LESSONS LEARNED

Periodic sampling of individual containers should be performed to validate the applicability of overall characterization performed on large waste streams. Sampling should be skewed to containers at the beginning and end of waste generating processes.

Physical processes during Decontamination and Decommissioning may lead to concentration of loose contamination as the project progresses.