The Remediation of Hanford's Last Low-Level Waste Burial Grounds in the 300 Area: 618-7 and 618-1 - 9487

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ABSTRACT

Under the U.S. Department of Energy's (DOE) River Corridor Closure Project, Washington Closure Hanford (WCH) has completed remediation of more than seven low-level waste (LLW) burial grounds in the 300 Area of the Hanford Site. The records of decision for the burial grounds required excavation, characterization, and transport of contaminated material to a *Resource Conservation and Recovery Act of 1976*-compliant hazardous waste landfill. This paper discusses the challenges and lessons learned from remediating the last two major burial grounds in the 300 Area: 618-7 and 618-1.

The 618-7 Burial Ground was in operation from 1960 through 1973, during which it received waste from the production of Zircaloy (zirconium alloy) jacketed metallic uranium fuel rods and thoria targets for the production of uranium-233. Its major remediation challenges included the recovery, characterization, and disposal of 550 drums and disposal of two compressed gas cylinders that were suspected to contain highly toxic chemicals. Approximately 100 of the drums contained Zircaloy metal turnings that could be pyrophoric under certain conditions. Remediation activities were completed in December 2008.

The 618-1 Burial Ground was in operation from 1945 (i.e., the beginning of Hanford operations) through 1951. It received waste from 300 Area laboratories that conducted experimental work associated with World War II and Cold War era processes for fuel fabrication and the production of plutonium. Some of the wastes were associated with highly radioactive irradiated material. Remediation of this burial ground is still in progress and is expected to be completed by June 2009.

Information presented in this paper will be an aid to those involved in the planning, design, and remediation of burial grounds located on the DOE complex.

INTRODUCTION

WCH is a limited liability corporation owned by Washington Group International, Bechtel National, and CH2M HILL selected to manage the $1.9 billion Hanford River Corridor Closure Contract (RCCC) for DOE. The RCCC work scope includes the deactivation and cocooning of plutonium production reactors, remediation of burial grounds and waste sites along the Columbia River, and the operation of the Environmental Restoration Disposal Facility (ERDF). In August 2008, WCH completed the third year of its 7-year contract.

This paper summarizes the remediation challenges and lessons learned from the remediation of the last two LLW burial grounds located in the 300 Area of the Hanford Site: 618-7 and 618-1 (Fig. 1). The remedial actions for these burial grounds was covered under a Record of Decision that required contaminated material above an industrial cleanup use level be removed and transported to a Resource Conservation and Recovery Act of 1976 compliant landfill located in the 200 Area of the Hanford Site.

Remediation of the 618-7 Burial Ground began in January 2008 and was completed in November 2008. Remediation of the 618-1 Burial Ground began in September 2008 and is scheduled to be completed by June 2009. This paper will assist engineers and regulators involved in the planning and remediation of LLW burial grounds within the DOE complex.

Fig. 1. Location of the 618-1 and 618-7 Burial Ground.

HANFORD BACKGROUND

A brief summary of Hanford's operations is provided for a better understanding of the nature and types of burial ground waste. The Hanford Site, located in Washington State, encompasses 1,517 km^2 (586 mi^2) and is divided into three major areas. The 100 Area, located at the north end of the site, contained nine plutonium production reactors. The 200 Area, located in the center of the site, contained the chemical processing facilities and the high-level waste storage tanks. The 300 Area, located at the south end of the site, contained the former fuel fabrication facilities and research laboratories.

The 100 Area contained nine production reactors operated between 1945 and 1986. Eight of these reactors were of an older design that used single-pass cooling water. The single pass reactors were phased out of operation by 1971. All of the single-pass reactors used aluminum jacketed fuel targets. Beginning in 1963, the more modern N Reactor was brought online. The N Reactor used a closed loop cooling system and a cylindrical target with a Zircaloy-2® metal jacket.

The 200 Area contained chemical processing facilities for removing plutonium from the irradiated reactor fuel. Irradiated fuel from the 100 Area was transported by rail car to chemical processing facilities in the 200 Area. Here the targets were stripped of their jackets and dissolved in an acid solution. The acid solution then underwent numerous chemical processing steps to separate plutonium from the unwanted fission products (e.g., strontium and cesium). The refined plutonium was then shipped offsite for fabrication in nuclear weapons components. The highly radioactive fission products were then sent to large underground storage tanks. The storage tanks are currently part of the Hanford Waste Treatment Program which will eventually immobilize the radioactive contents from the tanks into a vitrified glass.

The 300 Area was primarily used for research facilities and production of fuel targets for operation of the production reactor. Initially (1943), it housed the research facilities and fuel fabrication facilities required to support the construction of the first production reactor. It later evolved to contain numerous research facilities for plutonium refining, irradiated fuel examination, and radioisotope research. Between 1944 and 1957 more than 1,000 research tests were performed in the 300 Area [1]. Many of these tests produced unique waste that was sent to the 300 Area burial grounds.

The 300 Area fuel fabrication facilities produced fuel elements for the single-pass reactors and N Reactor. Single-pass fuel was produced between 1944 and 1971. Its production consisted of the heating and extrusion of uranium billets into rods, out-gassing and straightening of the rods, cutting and machining of the rods into elements, cleaning and degreasing the elements, cladding, and then inserting the elements in an aluminum jacket. On average, 30,000 fuel elements were produced weekly for the single-pass cooling reactors. [1]

N Reactor fuel was produced from 1961 to December 1986. It differed from single-pass fuel in that the fuel consisted of two concentric rods of fuel (an inner and outer element) that was co-extruded with the Zircaloy-2 cladding. The extrusion process was performed by placing a Zircoloy-2 inner and outer shell around a prepared uranium billet. This assembly was heated and then pressed through an extrusion die. The high pressure exerted during the extrusion process resulted in a strong bond between the cladding and the uranium metal. The extruded tubing was then cut to length, machined, and caps were welded over the ends to complete the fuel element. N Reactor fuel production reached a peak of 250 elements per week in the mid-1980s. [2]

618-7 BURIAL GROUND

The 618-7 Burial Ground, west of the Hanford Site 300 Area complex (as shown in Figure 1), was a general purpose burial ground operated between 1960 and 1973. The original burial ground consisted of one east-west-oriented trench, 198 m (650 ft) long, 30 m (100 ft) wide, and about 4 to 4.6 m (13 to 15 ft) deep. In 1965, a second trench, identical to the first trench, was constructed about 6 m (20 ft) north of the northern edge of the first trench and began receiving waste in 1966. At about the same time, a third trench was constructed to the south of and parallel to the original trench. This “V” shaped trench was 140 m (459 ft) long and 9.1 m (30 ft) wide. It received wastes from a research program on thorium target production.

Prior to remediation activities, WCH prepared an integrated hazard evaluation (IHE). The IHE summarized historical records in an effort to quantify the types and quantities of chemical and radiological material sent to a burial ground. It also reviewed the history of surrounding facilities in an effort to identify other waste streams that may have been sent to a burial ground. The IHE provides a

rigorous approach to ensure hazards are addressed in the site specific health and safety plan. A listing of radionuclides and contaminants from the IHE is shown in Table I.

Table I. 618-7 Burial Ground Contaminants from the Initial Hazard Evaluation.

Oxides		
Americium Oxide Powder	Hydrogen Peroxide	Plutonium Oxide
Cupric Oxide	Neptunium Oxide	Sodium Hydroxide
Curium Oxide Powders	Peroxide	Uranium Oxides
Acids		
Acetic Acid	Hydrofluoric Acid	Picric Acid
Aqua Regia	Nitric Acid	Sulfuric Acid
Boric Acid	Oxalic Acid	Tartaric Acid
Citric Acid	Phosphoric Acid	Magnesium Perchlorate
Metals		
Aluminum	Gadolinium	Mercury
Arsenic	Gold	Nickel
Beryllium	Iron	Praseodymium
Calcium	Lanthanum	Silver
Chromium	Lead	Sodium
Copper	Manganese	Zinc
Radionuclides		
Americium-241	Iodine-132	Ruthenium-103
Arsenic-76	Manganese-54	Ruthenium-106
Cerium-144	Manganese-56	Scandium-46
Cesium-137	Neptunium-237	Sodium-24
Chromium-51	Neptunium-239	Strontium-90
Cobalt-60	Phosphorus-32	Tantalum-182
Copper-64	Plutonium-238	Thorium-228
Curium-244	Plutonium-239	Thorium-232
Curium-252	Plutonium-240	Tritium (H-3)
Gallium-72	Plutonium-241	Uranium-235
Indium	Promethium-147	Uranium-238
Iodine-131	Radium-226	Zinc-65
		Zinc-69

Other Contaminant Groups (present but not listed)		
Nitrates	Molybdates	Chromates
Ketones	Sulfates	Iodides
Flourides	Phosphates	
Organics		
Ethanol	Graphite	
Normal Paraffin Hydrocarbon (Kerosene)	Dinitrophenol	
Picric Acid	Acenaphthene	
Metal Alloys		
Aluminum-Silicon	Beryllium-Zirconium	
Beryllium-Zircaloy-2	Sodium-Potassium	
Carcinogens		
Asbestos	Carbon Tetrachloride	Plutonium*

Cupferron	Benzene	Phenolphthalein*
Formalin	Trichloroethylene	Nickel Chloride*
Beryllium	Arsenic	Uranyl Nitrate Hexahydrate*
* Anticipated carcinogen		
Miscellaneous		
Ammonium Fluosilicate	Laboratory Hoods (20)	Radium-Beryllium Sources
Cell Cleansers	Metallic Dusts Fines	Reactor Fuel Elements
Chemical Reagents	Metallic Foils	RECUPLEX Process Waste
Concreted Drums of Pyrophoric Uranium	Methyl Orange	REDOX Process Waste
Construction Debris	Plutonium Waste Container Reading 5 Million D/M	Resins
Contaminated Construction Debris	Boron	Ruptured Fuel Slug
Drums of zirconium turnings >200	Polonium-Beryllium Sources	Ruptured Or Failed Fuel Rods Containing Plutonium And Fresh Fission Products.
Degreasers	Potassium Permanganate	Soil Contamination
Dibutyl Butyl Phosphonate	PUREX First Cycle Waste	Uranium (Chips And Dusts)
Ferrosulfamate	Pyrophoric Uranium Scrap	Uranium Slugs
Filters	Radioactive And Mixed Waste	Gloveboxes For Working With Radioactive Materials

The most notable concern identified in the IHE was that the burial ground contained hundreds of drums of Zircaloy-2 turnings from the fuel fabrication process. Similar to zirconium, Zircaloy-2 turnings may ignite at ambient temperature when exposed to air and are extremely difficult to extinguish once ignited. Due to the close proximity of this burial ground to a major highway providing access to the Hanford Site and the Columbia River, minimizing the potential of a drum fire was a major objective of the remediation. To address these concerns, WCH developed a comprehensive drum (or anomaly) handling program (Fig. 2).

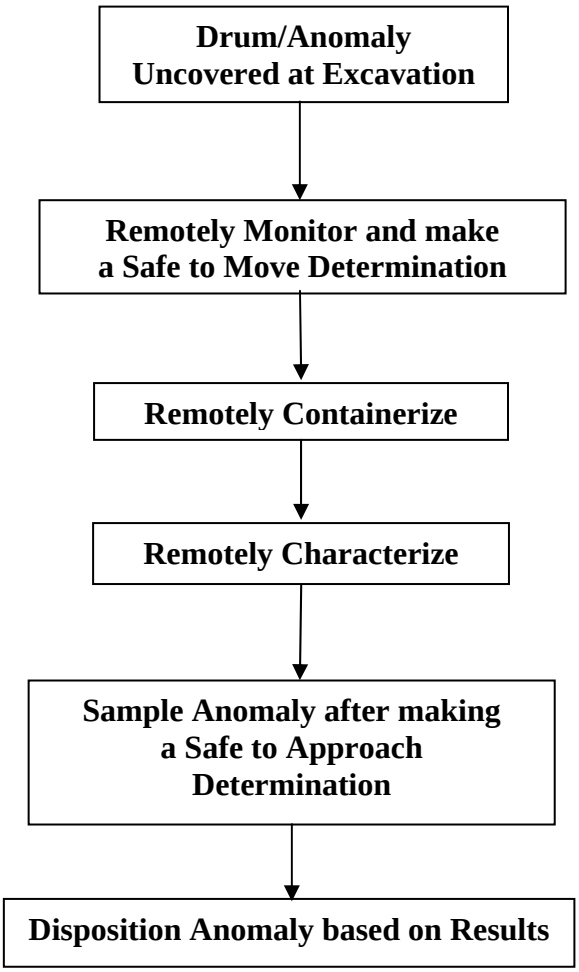

Fig. 2. Flow Chart for Handling Drums or Anomalies.

The drum handling program included administrative and engineering controls to minimize the potential for a fire. When a drum or anomaly was uncovered at the excavation it was remotely monitored for temperature, volatile organic compounds (VOCs), and radiological dose rate. The monitoring equipment was mounted on the boom of the excavator and could be directly positioned over a drum. The measurements were then evaluated by the health and safety officer and the radiological engineer before giving approval to move the drum. If approved, the drum was then containerized using an excavator. Using an excavator to containerize drums is a unique operation that was developed by a talented equipment operator working on the project. It greatly improved productivity and minimized risk by eliminating the need for personnel to wear protective clothing and work near an uncharacterized drum.

After a drum was containerized it was moved using a long reach forklift equipped with grapplers to a non-destructive characterization line. The purpose of the characterization line was to collect information to determine if a drum could be safely sampled. This information was collected remotely and included: weight, a gamma spectral analysis, and isotopic neutron spectroscopy.

The gamma spectral analysis was performed with a portable instrument that was able to quickly identity many radionuclides. The instrument was also able to differentiate between the various enrichment grades of uranium and plutonium. Isotopic neutron spectroscopy (INS) was used to help identify contents of drums without the need for opening. It can be used to identify elements like mercury, lead, and

zirconium. The effectiveness of INS for identifying drums containing zirconium was not fully validated because of equipment malfunctions.

Based on the non-destructive characterization results, a determination was made as to whether a drum should advance to the drum penetrating facility (Fig. 3). This facility provides a High-Efficiency Particulate Air controlled environment in which drums can be remotely opened and monitored before sampling. It is equipped with a video camera for monitoring operations and has instrumentation for monitoring temperature, VOCs, and β/γ radiation. In the event that a drum ignites, sand could be released from an overhead hopper to smother the fire. Research conducted for this project determined sand to be the preferred methods for extinguishing a zirconium fire.

A drum to be sampled was placed in the facility using a long reach forklift. The doors to the facility are then remotely closed. A pneumatic lift then raises the drum upwards until a brass punch punctures the lid of the drum. The lift is then retracted and the drum contents are inspected using a high quality video system. If the drum was suspected of containing zirconium turnings, water was then added via a nozzle attached to the punch mast. The drum is then monitored for a 2 minute period to verify that radiation and combustive gas levels are safe for sampling. At this point the doors are opened and sampling personnel can enter in Level B protective equipment.

Fig. 3. Drum penetrating facility used to open and sample drums and bottles.

Operation of the sampling facility proved so successful that it was modified to sample bottles which contained liquids or solids. The modification involved replacing the mast with a foot. A bottle to be sampled was placed inside a plastic tote container. The tote was then positioned under the foot. The pneumatic lift was raised until the bottle was crushed inside the tote. The contents of the bottle were then recovered for analytical analysis. As with drums, the entire process could be remotely monitored; thereby, limiting the potential exposure to samplers.

In total, over 550 drums (approximately 100 contained zirconium turnings, 200 contained aluminum turnings, 200 contained miscellaneous trash, and 50 contained oil) were processed through the drum

sampling facility without an event or safety concern. Most drums found in the burial ground were of a carbon steel construction, although a few stainless steel drums were recovered.

Other challenges involved with this remediation included discovery of unknown gas cylinders, spontaneous ignition of excavated material, controlling airborne uranium levels, and treatment of lead contaminated soil. During excavation operations, two compressed gas cylinders of unknown contents were discovered. Initial inspection indicated that the cylinders were still intact and potentially pressurized. One of the cylinders, which measured 10 cm (4 in.) in diameter by 0.5 m (20 in.) long, had a hexagonal shaped valve head the other was slightly smaller and had a rounded valve cover (valve not visible). A literature search of historical documents indicated these styles of cylinders most likely contained phosgene or hydrogen cyanide.

Due to the toxic nature of both gasses, special precautions were taken for their venting and treatment. WCH subcontracted to Integrated Environmental Services of Atlanta, GA, to vent and treat these gas cylinders. Both cylinders were transferred to a remote location at the Hanford Site in a U.S. Department of Transportation approved over-pack container for pressurized gas cylinders. The cylinders were then remotely vented in a containment vessel. Off-gas from the vessel was routed to an off-gas treatment system. Both cylinders were vented without incident and determined to contain nitrogen gas.

Towards the end of excavation activities in the northern trench, a pile of material erupted into flames. The equipment operator working nearby immediately covered the pile with soil. Work was stopped until an investigation could determine the cause of the fire. Sampling conducted in the area of the fire could not determine the cause or source of the fire. It was postulated that fire may have been the result of a broken bottle or reactive chemical disposed in the burial ground. Work was resumed in the area and work progressed without further incident. Unexpected small fires or flashes have occurred at several other burial grounds and should be anticipated.

An estimated 1,500 metric tons (2,000 US tons) of debris was size reduced so that it could be loaded into shipping containers (Fig. 4). Debris that was sheared included stainless tanks, piping, process equipment, and other types of construction waste. Air monitoring conducted around the shearing operations showed some of the highest airborne contamination readings recorded during the duration of the project. Most of the contamination was determined to be uranium and thought to be associated with the process equipment. To lessen contamination levels, the debris piles were sprayed with a fixative.

During excavation of the middle and north trenches, 18,000 metric tons (24,000 US tons) of soil failed the U.S. Environmental Protection Agency Toxicity Characteristic Leachate Procedure testing for lead. This material required solidification before it could be placed in the disposal landfill. Solidification was performed within the disposal landfill using a mixing box. Soil to be treated was placed in the mixing box along with Portland cement and water. The material was then stirred using the excavator bucket until fully mixed. It was then removed from the mixing box and pushed with a dozer into the disposal area. During peak operations, approximately forty 15 m^3 (20 yd^3) containers of soil were solidified per day. Solidification operations had to be regularly halted during high winds because the cement became airborne.

Lastly, the IHE for 618-7 identified the potential for beryllium contaminated waste. Due to the sensitivity of beryllium in the DOE complex, a conservative remediation approach was taken to meet the beryllium requirements in US Title 10 Code of Federal Regulation (10 CFR) Part 850. Meeting these requirements greatly increased remediation costs. It required items leaving beryllium regulated areas to be wiped down, monthly sampling to be conducted in eating and change areas, and additional training and medical surveillance for workers. Getting workers trained and certified for beryllium work alone was a

three week process. In the end, no airborne beryllium was found, although it was present in some waste and was naturally occurring in the soil.

Fig. 4. Metal Debris coated with Fixative prior to shearing.

618-1 BURIAL GROUND

The 618-1 Burial Ground (also known as Solid Waste Burial Ground No. 1, 318-1) is located in the northeast corner of the 300 Area (Fig. 1). It was active from 1945 through 1951 and, based upon geophysical investigation, consists of three trenches running north-south and measuring 5 m (16 ft) wide by 61 m (200 ft) long by 2.4 m (8 ft) deep and an associated neutralization pit located at its south end. The burial ground received waste from early 300 Area facility operations, including a test reactor and miscellaneous laboratories. The IHE identified uranium, plutonium, and fission products and potentially pyrophoric uranium oxide and metal chips.

Remediation work on this burial ground started in September 2008 with the excavation of the acid neutralization pit. The acid pit was found to contain contaminated soil and miscellaneous debris (e.g., steel vessels, drums, and bottles). As of January 2009, work is just beginning on the excavation of the first of its three trenches.

The burial ground is expected to contain 18,600 m^3 (24,300 yd^3) of contaminated soil, process equipment, building debris, and laboratory glassware. It will be remediated using techniques developed for the adjacent 618-2 Burial Ground which was remediated in 2006 and the drum sampling facility developed for the 618-7 Burial Ground.

The 618-2 Burial Ground is most noted for containing a locked combination safe which contained gram quantities of plutonium. A jug recovered from the safe was found to contain the world's oldest collection of plutonium-239 and is considered to be of historical significance [3]. The 618-2 remediation presented significant challenges related to the control of airborne alpha contamination. After an incident where two radiological control technicians received an uptake while taking a smear, the entire remediation process was revaluated. Several changes were implemented to prevent future contamination incidents. The most significant was applying magnesium chloride during excavation activities. The magnesium chloride, because it's hydroscopic and retains moisture, was found to minimize dusting of stockpiled soil even under arid summer conditions. Another change was made to the daily air monitoring program to better warn of changing radiological conditions. Air monitor filters were counted before the start of daily operations. If the air sampling results did not decay to the anticipated levels for naturally occurring radon, remediation operation did not resume until the results could be verified with alpha spectroscopy.

SUMMARY

Remediation of the 618-7 Burial Ground was completed in December 2008; the 618-1 Burial Ground is proceeding without incident and is expected to be completed in June of 2009. The completion of these burial grounds without a major safety incident is the result of experience gained from remediation of seven other 300 burial grounds. Some important lessons learned from these burial grounds include the following:

- Review historical records including radiological surveys to determine the types of waste that may be present in a burial ground. WCH summarizes this information in an IHE. The IHE is essential in designing a health and safety monitoring program that is protective of the workers. It ensures that a means is in place to monitor all contaminates, including those that may be hard to detect. The exposure incident at the 618-2 Burial Ground was the result of a hard to detect radionuclide not being seen during routine surveys and it resulted in an exposure incident and a year of down-time that more than doubled the length of the project.
- Anticipate that short term flash fires may occur and their source may never be determined. Workers and regulators should be briefed on emergency response actions to minimize costly down time.
- Be aware of the dangers associated with different types of compressed gas cylinders. The construction of the gas cylinder may give an indication that it contains a particularly toxic gas.
- Control of wind-blown dust and debris is essential to preventing airborne contamination releases. The application of a magnesium chloride solution was successful in controlling dust problem in the arid Hanford environment.
- If beryllium waste is present, evaluate its potential to become airborne. Meeting the beryllium requirements in 10 CFR Part 850 will result in cost and schedule impacts.
- If drums are encountered, consider training operators to over-pack drums remotely using an excavator. This method eliminates the need for workers to approach uncharacterized drums. In

addition, the need for a drum opening facility should be evaluated if drums contain pyrophoric material or other hazardous material.

- Evaluate unusual waste to determine if it may have historical significance, e.g., the jug recovered from the 618-2 Burial Ground was found to contain the oldest known collection of plutonium-239 in the world and is considered to be a significant discovery.

In conclusion, much knowledge has been gained from 10 years of burial ground remediation experience in the Hanford 300 Area. Many of the lessons learned discussed in this paper are applicable to other DOE sites and may result in significant time and cost savings if taken into consideration during a project's design and planning phases.

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