The Legacy of Nuclear Research
Waste Disposition Initiatives at Argonne National Laboratory, Argonne, IL – 10563

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ABSTRACT

Argonne National Laboratory (Argonne) has made many contributions in the field of nuclear fuel and reactor design, determination of the effects of radiation on materials, and basic nuclear physics throughout its 50-year history. As a result, Argonne wastes include most of the known radionuclides and many hazardous and uncommon chemical materials. Over the past few years, Argonne has ambitiously pursued the management of excess materials and equipment that supported past missions. Initiatives such as the Legacy Waste Project, Unneeded Materials and Chemicals Project, and Nuclear Footprint Reduction Project have made excellent progress to safely and compliantly characterizing, packaging, and dispositioning excess radioactive and non-radioactive wastes and materials. These projects are freeing up associated facility space for new research or supporting facility demolition for those facilities no longer needed. Special challenges have been and continue to be met in order to safely and compliantly characterize, package, and disposition wastes from Hazard Category 2 and 3 facilities, as well as radiological and non-radiological facilities.

INTRODUCTION

In 2007, Argonne kicked off three related but separate initiatives to address legacy wastes. The first was the Legacy Waste Project, established to address a population of problematic wastes remaining in permitted storage facilities. The second was the Unneeded Materials and Chemicals (UMC) Project, which was mandated by a September 1999 Department of Energy (DOE) Inspector General Audit Report, entitled "Management of Unneeded Materials and Chemicals (UMCs)," CR-B-99-02. The third was the Nuclear Footprint Reduction and Deactivation (NFRDP) Project, established in response to Argonne's decision to end programmatic nuclear research in Hazard Category 2 and 3 facilities.

Each of the above initiatives was needed to support continued operations at Argonne, but initial funding for them was less than required. All three were initially supported from existing Argonne funding; however, it was apparent that additional funds were needed to meet the schedule expectations for these efforts. In 2009, $29 million in additional appropriations was provided to Argonne specifically for the purpose of legacy nuclear waste and materials cleanup. These funds, which originated from the Department of Energy's (DOE's) Office of Environmental Management, Office of Science, and National Nuclear Security Administration, are managed as an overarching Omnibus project, which uses a work breakdown structure tied to specific facility areas for each of the Legacy, UMC, and NFRDP projects. Funding sources by facility are aligned with either the facility-specific historic liability or aligned with program-specific qualifying scope.

Highlights of the three (3) legacy waste cleanup initiatives at Argonne are provided below.

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LEGACY WASTE PROJECT

The Legacy Waste Project was initiated in January 2007 in response to identification of radioactive and non-radioactive wastes that had been stored at Argonne for up to twenty-five (25) years. Many of these wastes were not originally characterized in accordance with today's standards and, in some cases, were only characterized by a radiation survey. The initial volume identified at that time totaled approximately 10,000 cubic feet (ft^3), and included the following waste types:

- Low Level Radioactive Waste (LLW) - Residues, dry active waste, debris, aqueous and non-aqueous organic and inorganic liquids, sealed sources, filters, asbestos-containing material, remote handled waste
- Mixed Low Level Radioactive Waste (MLLW) - Combustibles, corrosives, reactive metals, elemental mercury, lead shapes, lead paint chips, miscellaneous lead debris, sludges, solvents, other aqueous and non-aqueous organic and inorganic liquids (Fig. 1)
- Polychlorinated Biphenyls (PCBs) - Articles, miscellaneous debris, oils
- Recoverable Material (RM) - Shield blocks, sealed sources, residues
- Remote- and Contact-Handled Transuranic Waste (RH- and CH-TRU) - Corrosives, other aqueous and non-aqueous liquids, debris

Disposition of vintage waste streams poses unique challenges, not all of which are technical. Regulatory concerns were first, i.e. addressing the MLLW that had been in storage for greater than the one year allowed under the Land Disposal Restrictions program. Argonne held discussions with the Environmental Protection Agency and subsequently submitted an updated Site Treatment Plan (STP) that included a schedule for these additional volumes of wastes. To date, the STP target dates have been met.

Fig. 1. Cabinet containing MLLW in RCRA-permitted storage area, and waste containers after packaging and loading for shipment to PermaFix.

Funding of the Legacy Waste Project was also a challenge. Argonne typically covers waste disposition costs through transfers from each generator's funding; however, the programs that

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had generated the above wastes had completed their missions and were no longer in existence. Prior to receipt of Omnibus bill funding for legacy waste cleanup, Argonne provided some funding internally from general funds. These funds were prioritized for use in addressing the MLLW.

The primary technical issue for the Legacy Waste Project has been the limited available characterization information and the worker exposure concerns associated with sampling higher hazard wastes. Former Argonne researchers were contacted to obtain additional process knowledge whenever possible, and real time radiography (RTR) and nondestructive assay (NDA) campaigns were implemented to supplement and/or confirm the process knowledge reviews (Figs. 2 and 3). Gamma-based NDA fell short of providing sufficient characterization for approximately 2,000 ft^3 of the M/LLW waste. For this subpopulation, the NDA analysis identified potential for TRU isotopes and, while other sources of information did not necessarily indicate potential for TRU isotopes, the available process knowledge was not strong enough to discount them. Argonne is in the process of designing and implementing physical sampling campaigns that minimize worker exposure. The time required for developing sampling and analysis plans, addressing analytical laboratory license limits, turnaround times for analytical results, and revising off-site facility waste profiles all contribute to the increased time it will take to disposition these wastes from the Laboratory, lengthening the process by approximately twelve (12) months.

Fig. 2. Examination of the physical form of drum contents using RTR unit.

Fig. 3. Analysis of drum contents using NDA.

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Obtaining compliant packaging has been a challenge for some waste items. For example, the CP5 reactor shield plug currently requires a Type B package (Fig. 4). This item was designed to provide a radiation shield at the top of the historical CP5 reactor, seal the reactor vessel, and provide vertical access into the vessel through several cylindrical penetrations in the plug. The plug, which weighs 27,700 lb, is 72 in. in diameter by 30 in. high. The exterior sides of the plug consist of ½-in. thick stainless steel and the bottom of 1-in. thick stainless steel plate. The stainless steel sides are lined with 3 ½-in. of poured lead, and the remainder of the plug is filled with high-density concrete. Activation radionuclide activity for the plug estimated at the time it was removed from the CP5 reactor (1997) was:

Co-60 5.4E+01 curies
Ni-63 6.30E+01 curies
Fe-55 5.00E+00 curies

Recent decay calculations have shown that within the next year, the activity of key nuclides in the plug will have been reduced by decay to levels permitting use of a Type A package. A suitable Type A package will be designed by a specialty packaging vendor to permit shipment of this high-activity mixed waste. Other challenging remote-handled wastes await similar analysis and packaging planning.

Fig. 4. Loading of the CP5 reactor shield plug into a storage cask.

TRU wastes are being organized into RH and CH TRU projects for incorporation into acceptable knowledge documents. Many of the legacy RH TRU containers include prohibited items and will require construction of engineering controls/structures to remove them. Many of the CH TRU wastes are acidic liquids that will be neutralized and solidified in processing facilities in the Waste Management Operations (WMO) Division.

The legacy RH-TRU wastes are characterized and certified in accordance with the central characterization program and, once approved, are scheduled and shipped to the Waste Isolation Pilot Plant (WIPP) consistent with an eight (8) week rolling schedule (Fig. 5).

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Fig. 5. RH-TRU drum being lifted/transferred from the gated drum shield.

The legacy CH TRU wastes will be shipped to Idaho National Laboratory (INL) under the Small Quantity Generator Program.

UNNEEDED MATERIALS AND CHEMICALS (UMCs) PROJECT

The driver for Argonne's UMC Project is the September 1999 DOE Inspector General Audit Report, entitled "Management of Unneeded Materials and Chemicals (UMCs)," CR-B-99-02, which cited DOE as a whole for not aggressively pursuing the disposition or reuse of UMCs, and stated that the DOE needed to strengthen its management of these items. The 1996 inventory, as estimated by the Inspector General, identified that there were at least 900,000 tons of such materials and chemicals within the DOE complex.

The UMC Project is being implemented in tandem with the Legacy Waste Project, and has followed a similar path regarding funding and technical issues. This project concerns legacy materials and chemicals that are no longer needed but are still located in research facilities and other locations rather than Argonne's permitted storage facilities. While it makes sense to retain these items for a certain period of time, they eventually decrease in value through time due to advancements in technology, expiration dates (e.g., chemicals), and refocusing of research efforts. Containers can degrade and result in leakage (Fig. 6), and some materials evolve into more hazardous substances, e.g., ethers evolving into shock-sensitive peroxides. Argonne has dovetailed the UMC cleanup efforts with programmatic changes designed to prevent future accumulation of these materials and chemicals beyond their useful life.

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Fig. 6. Example of unneeded chemicals in containers that deteriorated over time and breached.

The UMC Project focuses on materials and items that are inherently waste-like, abandoned, or speculatively accumulated, all of which meet the definition of "waste" under 40 CFR 262.11. A laboratory-wide inventory of UMCs was completed in 2007. A balanced approach is being used to disposition these materials, in which high-volume low-hazard items (e.g., discrete items that can be decontaminated, surveyed, and released as metals recycling moratorium waste) are expeditiously dispositioned in parallel with a smaller quantity of higher hazard items that require longer lead time for planning purposes.

Lower hazard items typically are dispositioned in campaigns. A semi-annual "clean sweep" is coordinated with Argonne's Building Managers to expedite removal of items from their facilities to reclaim valuable laboratory space, clear corridors, organize storage locations, and generally improve the operational and safety condition of site facilities. The Building Managers supply most of the basic labor resources and items, such as material release documentation, property management documentation, and operational coordination. The Facilities Maintenance and Services Division provides rigger support, as required, and WMO provides for recycle/reuse or waste disposal.

Chemicals (Fig. 7) are dispositioned primarily through "clearing house" locations. Once it has been determined that a chemical has no potential for on-site use, the owners transfer them to a central location within their building where a WMO crew, with support from a turnkey vendor, classifies, lab packs, and otherwise prepares them for off-site shipment to be recycled, treated, and/or disposed.

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Fig. 7. Examples of chemicals that are being collected from throughout laboratory facilities to be packaged for off-site disposition.

Oversized items amenable to size reduction are managed in campaigns (Fig. 8). An example of a campaign is the transfer of items that require size reduction to a yard area. Sizing, packaging, and off-site shipment are performed when sufficient quantities are gathered for efficient use of labor. This is very similar to the process used for disposition of chemicals but requires more up-front planning to have all the appropriate resources (e.g., riggers) and permissives (e.g., burn permit) in place.

Before Sizing

After Sizing

Fig. 8. Before and after photographs of a "Dempster Truck", which was size reduced and dispositioned under the UMC Project.

Disposition of other higher hazard items are managed as small projects. For example, a Zeus module no longer needed by the High Energy Physics Division was removed from service, packaged, and dispositioned off-site. The Zeus module was a 1,600-ton detector for measuring the energy and direction of particles with high precision (Fig. 9). It contained stacks of alternating plates of depleted uranium (7,588 kg) and scintillator plastic. In the center of Zeus, beams of protons collided head-on with beams of positrons (particles identical to electrons except for having a positive charge). Particles that sprayed out from the collisions interacted with the material in the detector. The plastic plates emitted ultraviolet light when struck by these particles, and fiber-optic cables transmitted the flashes to computers.

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Figure 9. Zeus module

A prototype cathode processor (PCP) currently installed in Building 308 has been identified as excess equipment by the Nuclear Engineering Division, and will be disassembled and shipped to INL for reuse (Fig. 10). The PCP is nearly identical to the cathode processor currently installed in the Fuel Conditioning Facility hot cell at INL, which is used in the treatment of spent sodium-bonded fuel.

The PCP provides a high-temperature vacuum furnace for research in processing spent reactor fuel, and can retort (distill) volatile components of the charge material and also produce a consolidated ingot of high-melting metallic components.

Figure 10. Prototype cathode processor installed in Building 308.

Cleanout of specific locations is managed as small projects that begin with removal of chemicals and packaging of loose items, followed by disassembly of installed components, then any necessary decontamination of the space, which may include removal of asbestos-containing components such as floor tile. These discrete areas are then available for reuse, or in some cases, demolition.

An example of a specific location that has been cleaned out is Chemical Sciences and Engineering (CSE) Division's Building 205, Room F-111, where researchers have been storing chemicals and equipment for potential reuse. Most of the chemicals were no longer useable (off-specification) or needed, and the equipment was out of date. Both CSE and WMO collaborated to develop a plan for safe cleanout of the area, beginning with radiation and industrial hygiene surveys to define potential hazards to workers. Larger low-hazard items (e.g., cadmium-contaminated hoods) were prioritized to be transferred out first to create more room to work with higher hazard containerized materials.

It was important to WMO that the Room F-111 materials be adequately characterized and packaged for shipment as they exited the facility to avoid the potential for having to handle the materials a second time. Process knowledge was used to characterize the wastes in conjunction with real-time analytical methods, such as radiation surveys and NDA, and when necessary, physical sampling and analysis. A Waste Certification Official (WCO) was present for packaging events, and verified that the inventory list for each package accurately reflected the contents, and that the contents met an Argonne waste profile for off-site disposal. While the WCO function is required only for shipment of LLW wastes to the Nevada Test Site (NTS), applying this independent oversight to MLLW resulted in documented quality assurance for meeting off-site waste acceptance criteria without additional visual inspection, RTR, or repackaging once the waste left Building 205. Most of the wastes from the Room F-111 cleanout have been dispositioned off-site (Fig. 11).

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Fig. 11. Before and after photographs of Building 205, Room F-111.

A second example of a UMC cleanout was completed at the Building 205 G-Wing laboratories. An initial sweep had been made in 2006 to disposition obvious waste items and expired chemicals that could be explosive (e.g., peroxide formers); however, lower hazard items still remained. To complete the cleanout this past year, prior to turnover for new research, each laboratory was reviewed for excess chemicals and/or waste material. CSE prepared the characterization documentation and submitted it to WMO for review. Once approved, WMO picked up the items and transferred them to WMO facilities for co-packaging with similar wastes and shipment off-site.

The on-site final packaging approach used at F-111 was particularly suited to the large volumes of waste to be packaged there. The smaller volumes of waste and diverse nature of waste at the G-Wing laboratories justified the approach to transfer waste to the central WMO facilities.

NUCLEAR FOOTPRINT REDUCTION AND DEACTIVATION PROJECT

In 2007, Argonne ended research and development activities at the existing Hazard Category (HC) 2 and 3 nuclear facilities. In concert with this decision, the NFRDP was established with the near-term mission of reducing inventories in each facility to less than the HC 3 limit. A graded approach is being used to develop documented safety analyses (DSAs) and nuclear Criticality Safety Evaluations (NCSEs) tailored for cleanup activities versus research activities, and this effort is being accompanied by facility upgrades as needed.

NFRDP Project objectives are summarized in the table below.

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Table I
NFRDP Objectives

	Reduce Inventory to <HC3	Submit Safe Harbor DSA with TSRs
FY2009	Vault 40 (complete)	Alpha Gamma Hot Cell (submitted)
FY2010	205 K-Wing	200 MA-Wing, 306, 331
FY2011	200 MA-Wing	Transportation Activities

De-inventory (removal of inventory above HC3) of Vault 40 has been completed. Materials and wastes were transferred to WMO HC2 storage facilities for an interim period, and have since begun shipping off site. Most materials have been shipped to the DOE Y12 Facility for reuse (Fig. 12); the wastes are being shipped to the NTS for disposal (Fig. 13).

Fig. 12. Preparation of Vault 40 Material for Shipment to Y12.

Fig. 13. Loading of Vault 40 Material for Shipment to the NTS.

The 205 K-Wing de-inventory will be completed in FY2010 (Fig. 14), including 1) encapsulation/packaging of fuel test specimen examination waste (FEW) currently residing outside the hot cells; 2) processing (neutralization and/or solidification) of liquids inside the hot cell; and 3) removal from inside the hot cell of FEW, residual solid material/debris, and processed and solidified solutions.

Fig. 14. Building 205 K-Wing first drum Visual Examination VE packaging operation

Processing, packaging, and removal activities for RH-TRU materials comprise the critical path to de-inventory Building 205 K-Wing to below HC3.

Solid material (including materials in hot cells when authorized) and waste removal activities will be ongoing throughout the de-inventory process. This category includes:

- processing equipment (some of which must be size-reduced);
- FEW currently in hot cells; and
- debris materials and waste.

The FEW material undergoes visual examination and is packaged as RH- or CH-TRU waste. The majority of the in-cell solid wastes will be characterized as RH-TRU waste, and the remainder will be CH-TRU or LLW.

The 200 MA/MB-Wing de-inventory will be completed in FY 2011. Initially, de-inventory activities will consist of removal of the material outside of the hot cell and the above-grade material within the hot cells. Work exterior to the hot cell will be performed as authorized in the implemented *Interim Safety Basis* (ISB) for Building 200 MA/MB-Wings. The controls in the ISB permit some non-routine maintenance and de-inventory of materials outside the hot cells in the facility, such as its Steel Room and Junior Cave (Fig. 15). The controls in the ISB also serve as the foundation for activity-specific hazard analyses, which have been submitted and approved by DOE.

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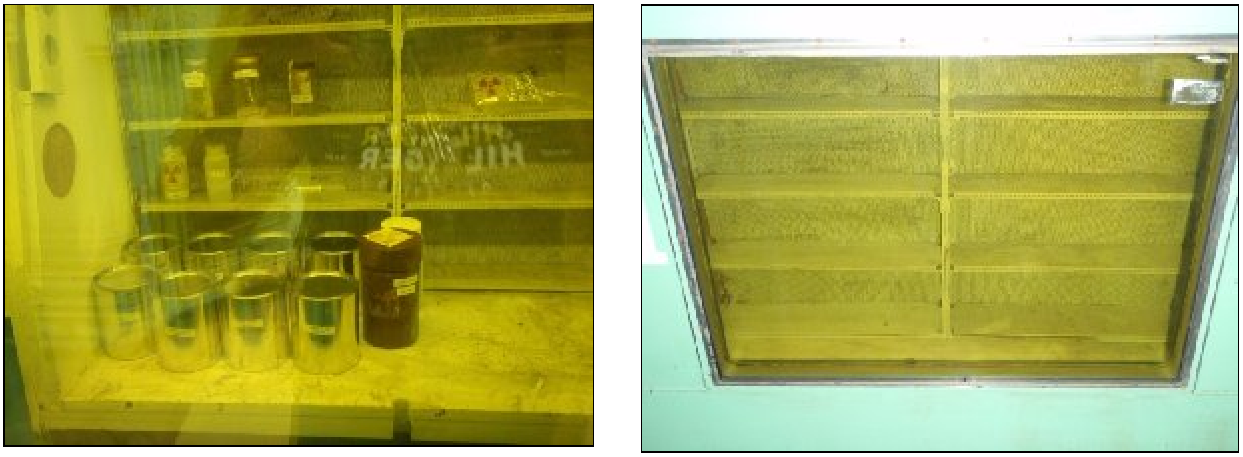

Fig. 15. Before and after photographs of the “Jr. Cave” in Building 200 MA/MB-Wing.