# Processing Plan for Potentially Reactive/Ignitable Remote Handled Transuranic Waste at the Idaho Cleanup Project - 12090

Patrick D. Troescher, Tammy L. Hobbes, and Scott A. Anderson
CH2M-WG Idaho, Idaho National Laboratory, Idaho Falls, Idaho 83415

## ABSTRACT

Remote Handle Transuranic (RH-TRU) Waste generated at Argonne National Laboratory – East, from the examination of irradiated and un-irradiated fuel pins and other reactor materials requires a detailed processing plan to ensure reactive/ignitable material is absent to meet WIPP Waste Acceptance Criteria prior to shipping and disposal.

## INTRODUCTION

One mission of the Idaho Cleanup Project (ICP) is to prepare remote-handled transuranic (RH-TRU) waste for disposal at the Waste Isolation Pilot Plant (WIPP). Facilities at the Idaho Nuclear Technology and Engineering Center (INTEC) have been modified to support repackaging, characterization, and shipment needs. The repackaging operation is designed to remove the waste contents from the original storage container; identify and remediate any WIPP prohibited items observed in the waste, and repackage the waste in to new drums. The new product drums containing repackaged waste are then further characterized for compliance with WIPP Waste Acceptance Criteria, certified, and prepared for shipment in the 72B cask. Waste characterization and certification operations are performed under the Washington TRU Solutions- Central Characterization Project (CCP) certification authority. Multiple RH-TRU waste streams at the Idaho National Laboratory (INL) have been qualified and approved for disposal at WIPP. Waste streams received from the Materials and Fuel Complex (MFC) have been further sub-divided into “lots” based on the generation location and attributes of the waste.

The Idaho Cleanup Project (ICP) received 48 canisters of waste for repackaging, characterization, and shipment to the Waste Isolation Pilot Plant (WIPP). These canisters were originally generated at Argonne National Laboratory-East (ANL-E), between November 1971 and November 1995 and stored at the Idaho National Laboratory (INL)-Materials and Fuel Complex (MFC) until 2009. In 2009 and 2010, these canisters were retrieved from MFC and sent to the INL Idaho Nuclear Technology and Engineering Center (INTEC) for repackaging and preparation for disposal at WIPP [1]. Existing hot cell facilities were renovated and placed into service to be able to safely process the RH TRU waste drums, while still being protective of the health and safety of the worker. This inventory of waste has been designated as “Lot 2” for purposes of grouping similar wastes for qualification for disposal at WIPP.

## BACKGROUND

The Lot 2 waste was primarily generated from post-irradiation destructive tests performed on a variety of fuels and other materials at ANL-E. The procedures of waste handling at ANL-E [2] segregated the debris into five categories as follows:

- Potentially Recoverable Fissile Material: “Discreet nuclear fuel residues resulting from post irradiation examinations, associated only with cladding materials and accurately identifiable as to composition and SPM batch number shall be considered potentially recoverable fissile materials."

- Nonrecoverable Fissile Material: Mixed fissile residues resulting from sectioning, grinding and polishing, and other operations which render the fissile debris unidentifiable as to the composition and SPM batch number shall be considered nonrecoverable fissile material. Fuel specimens in plastic metallographic mounts shall be included in this category and inventoried at one gram each. Fissile content shall be documented. Indefinite mixtures shall be inventoried according to procedures specified in the ANL Nuclear Materials Safeguards Manual.

- Contaminated Irradiated Structural Materials: Metallic and nonmetallic, noncombustible, nonfissile materials, which constituted cladding or other parts of fuel elements or experimental assemblies, which have undergone activation or cross contamination with activated materials shall constitute this category. Fissile content is to be documented if it exceeds 0.05% of the total mass.

- Noncombustible Operations Waste: Miscellaneous metallic and nonmetallic transuranic contaminated materials resulting from in-cell operations. Fissile content is to be documented if it exceeds 0.05% of the total mass.

- Combustible Operations Waste: Miscellaneous combustible transuranic contaminated materials resulting from in-cell operations. Fissile content is to be documented if it exceeds 0.05% of the total mass.

The segregated debris was placed in sealed metal containers (1-gal paint cans or equivalent)[2] and the contents inventoried. The contaminated irradiated structurals and noncombustible operations wastes were allowed to be combined for improved space utilization. Forbidden materials included liquids, halogenated plastics or organics, sulfur and sulfur compounds, sodium (except annular bonding sodium was permissible if plenum sodium was removed); and reactive chemicals. The cans were then loaded into the ANL-E canister for shipment to MFC.

Two thermal events occurred during repackaging of "Lot 2" RH-TRU waste. The first thermal event occurred at CPP-659 on August 30, 2010 during repackaging of ANL-E canister 11 in Can #189. This can contained Recoverable Fissile Material. The can contained both powdered fines and pieces of fuel. The most probable cause was identified as a metal fines fire due to the absence of characteristics typically associated with a sodium fire.

Repackaging operations for canister ANLE-11 were performed at the CPP-659 decontamination hot cell. Canister ANLE-11 contained a total of 8 inner 1-gal cans consisting of: two cans of non-combustible operations waste; five non-recoverable fissile material cans; and one recoverable fissile material can.

Approximately 23 minutes after Can #189 was vented, flames were observed coming from the top of the can vent hole (see Figure 2). Little smoke was observed coming from the fire. Met-L-X was poured onto the can and pushed into the can vent hole to extinguish the fire. The flame was also observed coming from the vent hole in the side of the can. The flames were extinguished approximately 2 minutes after being observed. Figure 3 shows the can after the flames were extinguished with Met-L-X.


Figure 1. ANLE-11 Can #189 Contents (Fuel and Powder).

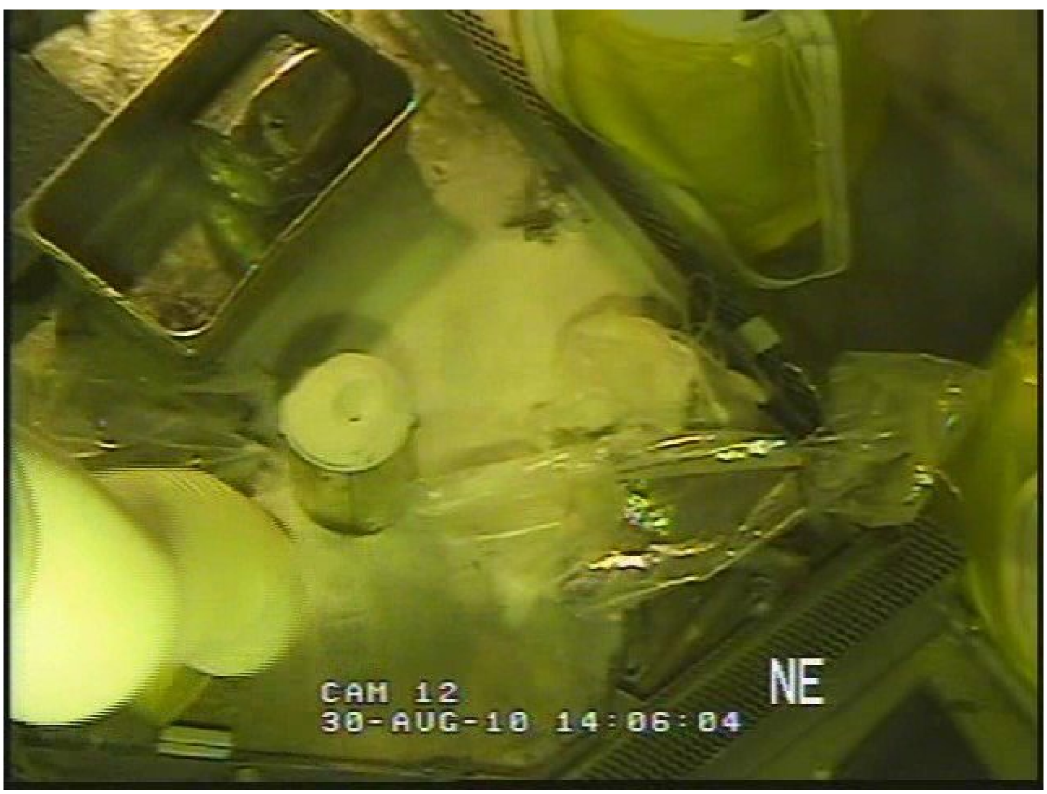


Figure 2. ANLE-11 Can #189 Fire During Event.


Figure 3. ANLE-11 Can #189 After Flames Extinguished with Met-L-X.

The second thermal event occurred at CPP-666 on September 13, 2010 during repackaging of ANL-E Canister 8 in Can #146. This can contained Recoverable Fissile Material. Can contents include EBR-II Mk-1A elements. Records indicated that this fuel was originally sodium bonded to the cladding. The AK stated that process for placing the fuel elements in the can included treatment of the sodium. No fines were observed in this can during the repackaging operation. The most probable cause of this event was identified as the presence of sodium in one or more of the fuel pieces.

Repackaging operations for ANLE-8 were performed at CPP-666 FDPA hot cell. The contents of canister ANLE-8 were eight 1-gal cans consisted of: one can of Recoverable Fissile Material with Bonded Clad Material (Can #146), one can of Non-Recoverable Fissile Material; three cans of Irradiated Structural Material; one can of Non-Combustible Operations waste; and two cans of Combustible Material.

Details on this event were obtained from a fact finding meeting and discussions with the operators involved with the repackaging operation. Can #146, which contained Recoverable Fissile Material, was observed as containing about 15-20 items (fuel pieces) approximately 5-inches in length and having the diameter and look of a “narrow screw.” Figure 4 shows the contents of the can. This “screw like” appearance is believed to be the wire wrap around the fuel piece. No fines were observed in Can #146. The can mass was reported as being approximately 4 lb. During removal of about 4 of the pieces from Can #146 to a new 1-gal can, sparks were observed as coming from the pieces. The sparks appeared red or orange-red. When these four pieces were dropped into the new can, there continued to be a glow and a whitish-grey smoke was observed. It appeared that several pieces were glowing in the bottom of the daughter can. Met-L-X extinguishing agent was added to the daughter 1-gal can. Observation of the items in Can #146 did not indicate any heat discoloration on the bottom of this can. Met-L-X was also added to this can. Each of the 1-gal cans (parent and daughter) resulting from repackaging Can #146 were individually placed in a 5-gal bucket that was filled with Met-L-X and the lid placed on the bucket (see Figure 5). Each 5-gal bucket was then placed in a 30-gal drum. The 30-gal drum was then overpacked into a 55-gal drum. Both drums were placed in permitted storage.


Figure 4. Can #146 Fuel Pieces.

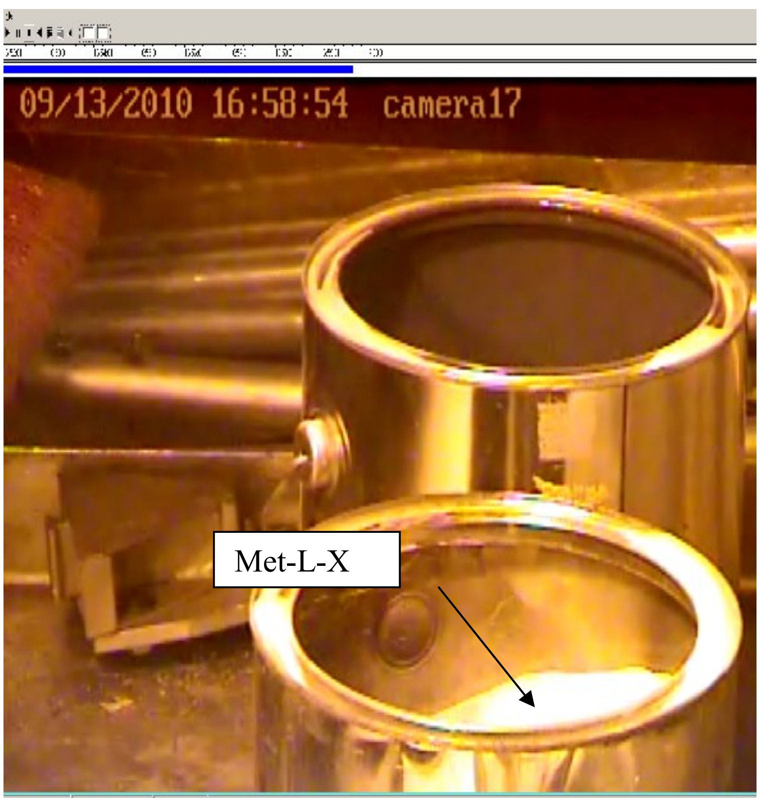


Figure 5. Daughter Can After Second Thermal Event with Met-L-X.

## DISCUSSION

As a result of the thermal events, there are two issues that need to be addressed. The potential for sodium metal reaction and the potential for ignitability event from the metal fines present during waste repackaging.

### Reactivity

A variety of fuel specimens were examined at the ANL-E hot cells. The majority of examined pins (see Figure 6) were from the Fast Breeder Reactor Fuel Program. Some fuels included sodium- or sodium-potassium- bonding to improve heat transfer properties. Irradiation of the fuel specimens included in Lot 2 spanned the time period of 1962 to 1983.

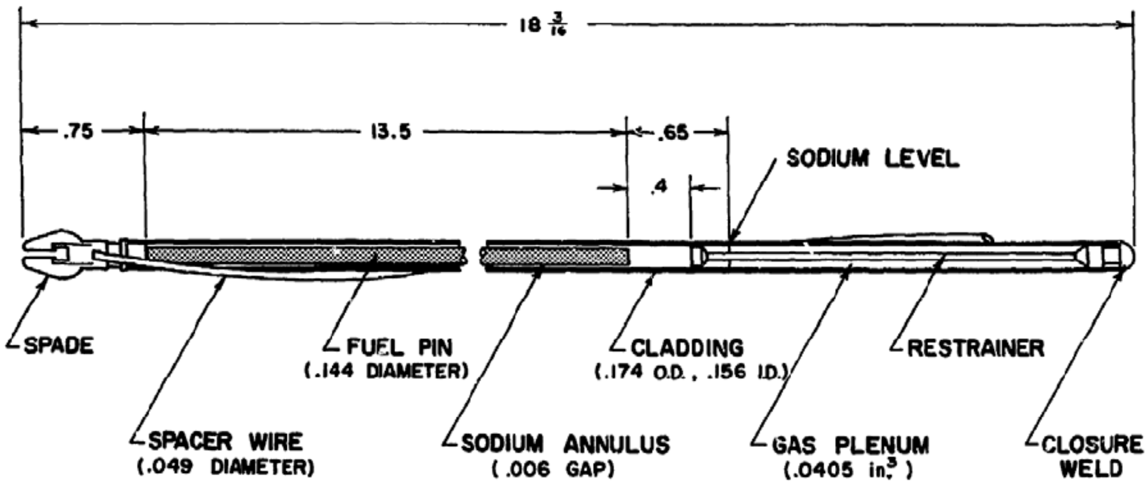


Figure 6. Experimental Breeder Reactor fuel pin.

Information (acceptable knowledge) collected from the waste generator indicates that residue from approximately 621 irradiated specimens were included in the Lot 2 waste. Of these specimens, approximately 225 specimens (36%) have been identified as being a sodium-bonded fuel.

Sodium-bonded fuel is prepared in roughly the same manner independent of the specific fuel element configuration. A measured amount of sodium is placed into a tube that has a fitting welded on one end. The tube will serve as the fuel cladding. The tube, containing the sodium, is heated in a furnace under an inert atmosphere. The tube is removed from the furnace, allowed to cool and the fuel rod is then inserted into the tube. The tube is placed into another furnace, usually with several other assemblies and heated just above the melting temperature of the sodium. The assemblies are vibrated during this heating process which settles the fuel rod and causes the sodium to be displaced into the annulus between the fuel rod and the tube (cladding). The top cap are inserted and welded closed. The tube is leak checked and then heated at a high temperature for several hours while being vibrated again. This process uniformly distributes the sodium in the assembly and centers the fuel rod within the assembly.

During preparation of the fuel specimen for the original destructive examination at ANL-E, sodium contained in the plenum area of the fuel pin was drained and reacted with alcohol. The fuel region of pin was then sectioned, typically multiple times, for further examination and testing. If cladding was removed from a sodium-bonded fuel, the sodium contained in the annulus area was also reacted with alcohol. Some of the fuel pin segments in the Lot 2 waste are expected to have intact cladding.

The ANL-E “Procedure for High Gamma Level Transuranic Waste Handling at the Alpha Gamma Hot Cell Facility”, MSD Document No. M1002-0047-SP-00, December 30, 1974 [2], provided specific guidance for packaging waste. This procedure specifically prohibited sodium (Na), potassium (K), or NaK in the waste except for sodium contained in the annulus area of the fuel pin (area between the outer diameter of fuel and inner diameter of cladding).

The annulus of sodium bonded fuels ranged from 0.006-inches to approximately 0.017-inches thick and would typically represent 0.6 – 1.7 grams of sodium in the annulus of the fuel zone, which generally was 13.5-inches in length. Fuel pins were subsequently sectioned for examination and individual sections or pieces of the fuel zone would only be expected to contain fractions of a gram of sodium. The fuel and fuel-like pieces along with other fuel examination wastes were packaged in an inert hot cell.

Since WIPP cannot accept waste with a D003 (reactive) hazardous waste number, further evaluation of the fuel and fuel-like pieces is necessary to verify that these items meet WIPP acceptance criteria. Calculations and tests have been recently performed by ICP on the fuel and fuel-like pieces. The calculations indicate that the amount of sodium present in each of the fuel pins, and an aggregate of several fuel pins was not sufficient to pose a hazard.

Several water immersion tests were conducted using unpassivated surrogate Mark 1A fuel pins. The surrogate pins were sectioned to expose the sodium bonding and placed back into an inert gas atmosphere. The use of freshly cut pins provided a worst case scenario as the sodium would be the most reactive. The pieces of the fuel pins were immersed in deionized water and the reactions were observed. The reactions consisted of the generation of very fine streams of gas bubbles, presumably hydrogen. The amount of hydrogen gas generated by these reactions is very small. The reactions were not vigorous, and, therefore, did not meet the regulatory definition of reactivity.

Tests were then repeated using fuel-like pieces that had been passivated by several different means (i.e., exposure to air, misting the exposed sodium bonding with water, and reaction with carbon dioxide. Moist carbon dioxide was used to passivate sodium in some of the coolant systems at EBR-II.) These tests were meant to address any concerns that the passivation of the exposed sodium surface might inhibit the reaction with water. The tests proceeded in the fashion as previous tests except that the reaction was initially slower after being placed in the water. After a short period of time the reaction progressed as in the unpassivated tests. The various types of passivation did not inhibit the reaction of the unpassivated sodium in the surrogate fuel pins. Again, the amount of hydrogen gas generated by these reactions is very small. The reactions were still not vigorous, and, therefore, did not meet the regulatory definition of reactivity.

## Ignitability

Fuel pins are made up primarily of uranium or UO2 fuel with a thin layer of sodium (or NaK) annulus and Zircaloy cladding (Figure 1). Zircaloy varies in composition but is always at least 95% Zr by mass. Cutting the fuel pins exposes the alkali metals, which, by their violent chemical reactions with water, are reactive according to RCRA, and therefore prohibited at the WIPP. The powdered swarf will be made up mostly of uranium or UO2, with small amounts of Na and zirconium. The trace amounts of Na in the swarf will oxidize in air without any violent reactions, but both uranium and zirconium, when finely divided, are ignitable. The high FGE content results from the high proportion of uranium in the waste.

Metals readily react with oxygen in air, as in the reaction:

$$U + O_2 \rightarrow UO_2$$

Because the oxidized metal compound is at a lower energy state than the pure metal plus oxygen, the reaction releases energy (heat). If this reaction occurs quickly enough, as when there is high surface area such as from metal fines, sufficient heat can be generated to reach the metal's ignition temperature. In many cases, metal shavings are stored in water (more than 25% by weight) to prevent ignition, a solution that is not plausible in this case. Furthermore, any treatment involving solutions is not possible due to the prohibition of liquid in the hot cells.

Another common method to eliminate the flammability of metals is to calcine them and treat the resulting (fully oxidized) clinkers as waste. The powder would need to be heated at a temperature above the metals' ignition temperature for 2 hours to ensure complete oxidation. A heating temperature of 800 °C is recommended for the uranium and Zircaloy fines present in the 48 canisters.

Another treatment method is to combine the uranium and zirconium metal fines with an inert ignition suppressing material, such as dry Met-L-X. A ratio of at least 5 parts Met-L-X to 1 part metal fines by weight is necessary to stabilize the swarf. Isolating the fine metal particulates by dispersing them throughout a suppressant prevents sufficient heat to develop during oxidation to lead to combustion [3].

## CONCLUSION

The Idaho Cleanup Project (ICP) approach to repackaging Lot 2 waste and how we ensure prohibited materials are not present in waste intended for disposal at Waste Isolation Pilot Plant "WIPP." uses an Argon Repackaging Station (ARS), which provides an inert gas blanket [4].

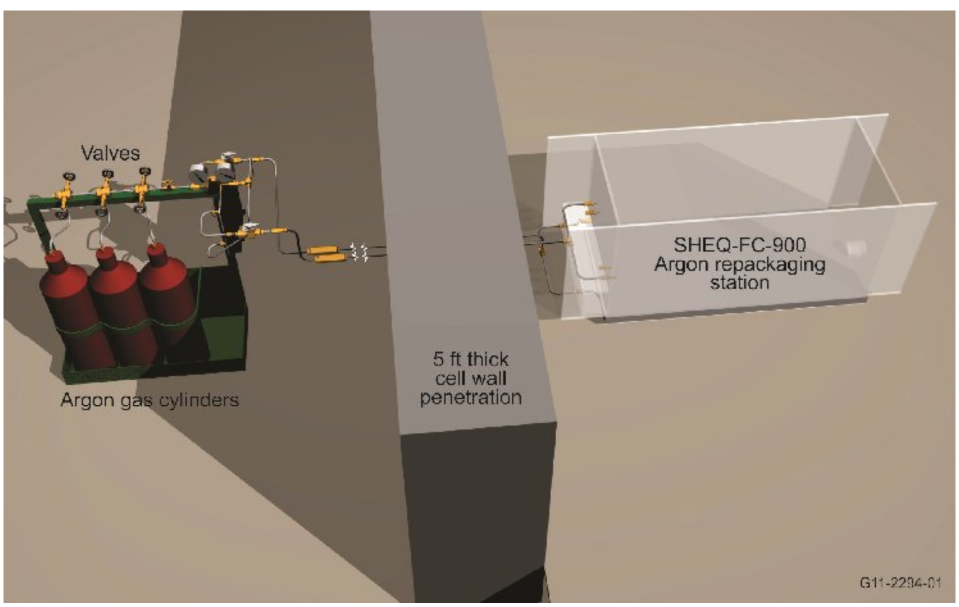


Figure 7. Argon Repackaging Station Assembly.

Opening of the Lot 2 containers under an argon gas blanket is proposed to be completed in the ARS. The ARS is an interim transition repackaging station that provides a mitigation technique to reduce the chances of a reoccurrence of a thermal event prior to rendering the waste “Safe.”

The consequences, should another thermal event be encountered, (which is likely) is to package the waste, apply the reactive and or ignitable codes to the container, and store until the future treatment permit and process are available. This is the same disposition that the two earlier containers in the “Thermal Events” were assigned. By performing the initial handling under an inert gas blanket, the waste can sorted and segregate the fines and add the Met-L-X to minimize risk before it is exposed to air.

The 1-gal cans that are inside the ANL-E canister will be removed and each can is moved to the ARS for repackaging. In the ARS, the 1-gal can is opened in the inerted environment. The contained waste is sorted, weighed, and visually examined for non compliant items such as unvented aerosol cans and liquids. The contents of the paint cans are transferred into a sieve and manipulated to allow the fines, if any, to be separated into the tray below. The fines are weighed and then blended with a minimum 5:1 mix of Met-L-X. Other debris materials found are segregated from the cans into containers for later packaging.

Recoverable fissile waste material (Fuel and fuel-like pieces) suspected of containing sodium bonded pieces) are segregated and will remain in the sieve or transferred to a similar immersion basket in the ARS. The fuel like pieces will be placed into a container with sufficient water to cover the recoverable fissile waste. If a “reactive characteristic” is present the operator will be able to observe the formation of “violent” hydrogen gas bubbles. When sodium bonded fuel-like pieces are placed in water the expected reaction is a non-violent reaction that does not meet the definition of reactivity. It is expected that there will be a visible small stream of bubbles present if there is any sodium-bonded fuel-like piece placed in the water. The test will be completed when there is no reaction or the expected reaction is observed. .At that point, the fuel like pieces complete the processing cycle in preparation for characterization and shipment to WIPP.

If a violent reaction occurs, the fuel-like pieces will be removed from the water, split into the required fissile material content, placed into a screened basket in a 1 gallon drum and drummed out of the hot cell with appropriate RCRA codes applied and placed into storage until sodium treatment is available. These “violent” reactions will be evidenced by gas bubbles being evolved at the specimen surface where sodium metal is present. The operators will be trained to determine if the reaction is “violent” or “mild”. If a “violent” reaction occurs, the sieve will be immediately removed from the water, placed in a 1 gallon paint can, canned in the argon cover gas and removed from the hot cell to await a future treatment. If the reaction is “mild”, the sieve will then be removed from the water; the material weighed for final packaging and allowed to dry by air exposure [5].

Lot 2 waste cans can be opened, sorted, processed, and weighed while mitigating the potential of thermal events that could occur prior to exposing to air. Exposure to air is a WIPP compliance step demonstrating the absence of reactive or ignitable characteristics.

## REFERENCES

1. CCP-AK-INL-500, Central Characterization Project Acceptable Knowledge Summary Report for Remote-Handled Transuranic Debris Waste from Argonne National Laboratory-East Stored at the Idaho National Laboratory.
2. Procedure for High Gamma Level Transuranic Waste Handling at the Alpha-Gamma Hot Cell Facility, MSD, ANL-E; MSD Document No. M1002-0047-SP-00; December 30, 1974.
3. Ams B., (2010), Recommendation for Treatment Path of 48 RH-TRU Canisters in Waste Stream ID-ANLES-5000, Los Alamos National Laboratory, NM.
4. Clements T.L. (2011), Probable Cause for Lot 2 Thermal Events, EDF-9982.
5. Sherick M.S, (2011), Argon Repackaging Station (ARS) Process Description, EDF-10025.