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EVALUATION OF FIRE HAZARDS WHILE REPACKAGING PLUTONIUM-CONTAMINATED SCRAP IN HB-LINE

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1. Summary

The potential for a fire while repackaging plutonium-contaminated scrap was evaluated. The surface-to-mass ratio indicates the metal alone will not spontaneously ignite. Uranium hydride can form when uranium metal is exposed to water vapor or hydrogen; uranium hydride reacts rapidly and energetically with atmospheric oxygen. The plutonium-contaminated scrap has been inside containers qualified for shipping, and these containers are leak-tight. The rate of diffusion of water vapor through the seals is small, and the radiolytic hydrogen generation rate is low. Radiography of samples of the storage containers indicates no loose oxide/hydride powder has collected in the storage containers to date. The frequency of a fire while repackaging the plutonium-contaminated scrap is extremely unlikely.

2. Introduction

HB-Line will receive plutonium-contaminated scrap, repackage the material in dissolvable bags and cans, and transfer the material to H-Canyon for dissolution. This material originated off-site. Loose corrosion products which were present on the surface of this material was removed by brushing before the scrap was packaged for shipment. After brushing, the material was first bagged in a polyvinyl chloride bag (during glovebox bagout) closed by taping. A polyethylene bag was then placed over the polyvinyl chloride bag and taped shut. The bagged scrap was then placed in an inner can with a screw lid (using a vulcanized silicone rubber gasket).. This inner can was placed in a leak-tight shipping container and shipped to SRS.

After arrival at SRS, the inner can was removed from the original package and placed in a 12-inch diameter sealed pipe component, which was subsequently placed in a Type 17C 55-gallon drum. It will remain in this configuration until shipped to HB-Line for repackaging.

Details of the operations to be performed in HB-Line are included in Appendix A. In summary, the inner can will be removed from the pipe overpack container and pipe component and transferred to a hood. The double-bagged plutonium-contaminated scrap will be removed from the inner can in a hood. The double-bagged scrap will then be passed into a glovebox and removed from the plastic bags. The scrap will then be bagged from the glovebox through a bagout port, and placed in a dissolvable can. The dissolvable can will be transferred to H-Canyon for dissolution.

Evaluation of other hazards will be performed in separate documents; for example, the hazards of inadvertent criticality will be evaluated in the Double Contingency Analysis (DCA). This report will only evaluate fire potential during the new operation.

3.0 Methods of Analysis

3.1 Plutonium-Contaminated Scrap Fire during Repackaging

Uranium metal, like several other metals, can be pyrophoric if it is in a finely divided form such as machining chips or dust from sawing (Reference 1). There has been a reported instance of uranium foil igniting when a bottle containing this foil was opened (Reference 2). However, fires of large pieces of uranium metal are not expected to occur except under the continuous addition of heat. No mechanical or chemical operations that might raise the temperature of the scrap are planned. The metal geometry will be evaluated to determine if the plutonium-contaminated scrap will be susceptible to ignition while repackaging. A fire from other sources could ignite the scrap, and these fires are judged credible. The consequences of burning plutonium-contaminated scrap will be evaluated relative to other missions authorized for HB-Line Phase I.

3.2 Uranium Hydride Reaction During Repacking

Uranium hydride is reactive with air, and large amounts of uranium hydride could react energetically when exposed to air. The mechanisms for uranium hydride formation will be examined, and factors which limit its formation will be identified and evaluated. A bounding analysis will be performed, and judgment will be applied when evaluating the probability of formation of large amounts of uranium hydride.

4.0 Results

4.1 Evaluation of Metal Fire

Peacock (Reference 3) has investigated self-ignition of uranium metal, and has correlated the susceptibility of self-ignition in air to the specific surface area (square centimeters/gram) of the material. Figure 3 of Reference 8 summarizes the information. Items to the right of this curve are not expected to self-ignite. Based on its surface to mass ratio, the plutonium-contaminated scrap is not expected to spontaneously ignite at room temperature.

The plutonium-contaminated scrap could be ignited by a fire in the glovebox or a full facility fire in HB-Line. Uranium metal burns when ignited, and generates heat at a slow rate. The RF/ARF for burning metal is 2.5E-04 (Reference 4, page 4-2). This is lower than the RF/ARF of 7.6E-03 used for boiling solutions of plutonium nitrite (Reference 5). Further, the dose conversion factors for inhalation for uranium (excepting U-232, which is not present in the plutonium-contaminated scrap) is less than for plutonium (Reference 6). Therefore, the doses calculated for processing 12 kilograms of plutonium oxide in a Mixed Scrap line bounds the doses from processing an equal amount of uranium for fire.

4.2 Evaluation of Uranium Hydride Fire

Uranium hydride reacts readily and energetically with oxygen (Reference 7). At least one pyrophoric event involving uranium metal corrosion products has been reported where uranium hydride was assumed to be the active agent (Reference 8). Unlike the plutonium-contaminated scrap storage, this event involved uranium metal coated with cladding and the cladding created a crevice environment that promotes uranium hydride formation.

Water vapor (and also liquid water) will react with uranium metal to form UO_2 and H_2 per the following reaction:

$$U + 2H_2O \rightarrow UO_2 + 2H_2 \qquad \text{Equation 1}$$

If the hydrogen is all contained in the vicinity of the metal surface (very conservative assumption), the hydrogen can react with the uranium metal to form uranium hydride, as shown in Equation 2.

$$2U + 3H_2 \rightarrow 2UH_3 \qquad \text{Equation 2}$$

Uranium hydride can be a finely divided powder, and reacts rapidly when exposed to oxygen. The reaction is shown in Equation 3.

$$4UH_3 + 7\,O_2 \rightarrow 4UO_2 + 3H_2O \qquad \text{Equation 3}$$

This reaction produced 1386 Kilojoules of heat per mole of uranium hydride (Reference 13).

Both the history of this material and its current storage conditions minimize the amount of uranium hydride that could be present during repackaging. This is discussed further below.

The plutonium-contaminated scrap is stored inside a PVC bag surrounded by a polyethylene bag (Reference 9) in a screw-lid can (SLC). There is a gasket on the screw-lid can to prevent particulate or liquid from entering or exiting the can, but gases and water vapor can diffuse into and out of the can through the vulcanized silicone rubber gasket.

The screw-lid can is placed in a 12-inch Pipe Component (PC). This PC is constructed from 12-inch Schedule 20 pipe. A cap is welded to the bottom of the PC, and a one-inch flange is welded to the top of the PC (Reference 10). The flange has an o-ring seal; the o-ring is Parker Seals Compound E603-70 Ethylene Propylene Size No. 2-382 (Reference 11). The opening for the 1-inch diameter sintered-stainless steel medium High-

Efficiency Particulate Air (HEPA) filter in a stainless-steel housing in the PC flange was plugged for PCs used for transporting and storing this material.

The PC was verified leak-tight by test (Reference 12) at the Rocky Flats Environmental Technology Site. The filter on the pipe was plugged, a port for evacuating the air from the pipe was prepared, and an air-tight plastic bag was placed around the pipe. The pipe was then evacuated, and the bag filled with helium. Atmospheric pressure was about 80 kilopascals, and the pressure inside the pipe was close to zero, so there was a differential pressure of helium across the pipe boundary of about 81 kilopascals (about 609 inches of mercury). The resulting inleakage was less than or equal to 1E-07 cm^3/second.

When initially loaded, the Pu-contaminated scrap was not loaded into a POC, but was loaded into a container of equivalent or lesser inleakage characteristics. Assuming the scrap has been in a POC container since being packaged for shipment is bounding for evaluating water vapor diffusion into the container.

The inleakage measured in the helium test described above is assumed to be principally diffusion through the o-ring. The permeability of the EPR o-ring, for helium at 25 °C (Reference 13), is 19.7 std cc·cm/(cm^2·second·bar).

The permeability of water vapor through the EPR o-ring, also from Reference 25, is given as 550 to 3700 std cc·cm/(cm^2·second·bar) at room temperature. This is a wide range. To compare the permeability of water vapor to helium, the extreme value of 3700 std cc·cm/(cm^2·second·bar) will be conservatively used.

The ratio of the rate of diffusion of water vapor to helium into the pipe overpack depends on the ratio of the permeability and of the driving force (partial pressure differentials). The partial pressure differential for helium diffusion is about 81 kilopascals, as stated above. The material is stored inside, but water vapor diffusion will be conservatively evaluated at outside humidity conditions. The 30-year average temperature and humidity at SRS is 64.7 °F and 69%, respectively (Reference 14). At 66 °F, the partial pressure of water is 0.316 psia (Reference 15). At 69% humidity, the partial pressure of water is (0.69)(0.316) ~ 0.218 psia, or ~ 1.5 Kilopascals.

The permeability ratio of water vapor to helium is (3700/19.7) ~ 188. The driving force ratio of water vapor to helium used in the test is (1.5/81) ~ 0.019. The diffusion ratio is ~ (0.019*188) ~ 3.6.

The rate of helium diffusion, as measured in the leak rate test, is less than or equal to 1E-07 cm^3/second. This is a yearly rate of 3.15 cm^3. So, the rate of water vapor diffusion, using very conservative assumptions for the permeability of water vapor in ethylene-propylene rubber, is (3.6*3.15) ~ 11.3 cm^3/year.

The maximum anticipated time the scrap would remain in this container is 9 years. The amount of water vapor that could diffuse into the pipe overpack, using the very conservative assumptions stated above, is (9*11.3) ~ 102 cm^3. For evaluation purposes, the hydrogen production from uranium reaction with water vapor will be rounded up to 110 cm^3.

The water vapor existed at an average temperature of 64.7 °F (291.1 °K). Using the Ideal Gas Law, and assuming one atmosphere pressure, the amount of water vapor that could react with the uranium metal is ~ (110/(291.1*82.06)) ~ 0.005 gram-moles. Per Equation 1, this would create 0.005 gram-moles of H_2. Theoretically, this could react to create ~0.003 gram-moles of UH_3.

A larger source of water vapor is the water vapor inside the PCV and screw-lid can when the material was originally packaged for shipment and then repacked at SRS. By measurement, the screw-lid container (SLC) is about 11.75 inches ID by about 11.75 inches high. The volume of the screw-lid container is about (0.785*(11.75)^2*11.75) ~1.273E+03 in^3 ~ 20.9 liters. The inside volume of the PCV is about (0.785*(12.725)^2*25.6) ~ 3.524E+03 in3 ~ 53.3 liters (Reference 32). The "free" volume in the PC is (53.3-20.9) ~ 32.4 liters. The average temperature and relative humidity at SRS is 64.7 F and 69 % (Reference 26). Conservatively assuming the PCV and SLC were at 64.7 °F (18.2 °C) when filled, the "free" volume of the PC contained (32.4/(291.2*0.08206)) ~ 1.36 gmoles of air, and the SLC contained (20.9/(291.2*0.08206)) ~ 0.87 gmoles of air.

Soon after arriving at SRS, the SLC was transferred to a PC, so the air in the PC was replenished.

It is conservatively assumed that 1/4 of the water vapor in the free volume of the shipper diffused into the SLC before transfer. This is conservative, since the plutonium-contaminated scrap is double-bagged as described above and the SLC has a vulcanized silicone rubber gasket (Reference 16) to prevent inleakage. Water vapor will diffuse through this gasket, but there must be a concentration differential between the water vapor in the PC and the SLC for this to occur. Since both bags containing the plutonium-contaminated scrap were taped (not heat-sealed), and there is a large surface area for diffusion, there will be significantly faster movement of water vapor from the SLC atmosphere through the bags than through the screw-lid gasket. There had not been time for full depletion of the water vapor in the SLC before the transfer was made.

The transfer to the PC was made indoors. The humidity of outside air was conservatively assumed for the building when evaluating water vapor interaction with the scrap.

Water vapor from the air in a PC, plus ¼ of the water vapor in the air from a PCV (during shipping), plus water vapor from the SLC is conservatively assumed to react with the plutonium-contaminated scrap. This is certainly a conservative assumption, since it assumes all the water vapor in the PC will diffuse into the SLC, and all the water vapor in the SLC will penetrate the double bags surrounding the scrap and the oxide coating on the scrap. The amount of air available is (1.25*1.36 + 0.87) ~ 2.57 gmoles. The amount of water vapor in this amount of air is (0.0148*2.57) ~ 0.038 gmoles.

Previously, a bounding value of 0.005 gram-moles of water vapor was calculated for diffusion of water vapor through the o-ring gasket in the PC. Summing the two amounts, the total gram-moles of water that could react with the scrap is (0.038 + 0.005) ~ 0.043 gram-moles.

Per Equation 1, one mole of water vapor generates one mole of hydrogen when it reacts with uranium. The amount of hydrogen generated is (0.043*82.07*291.2) ~ 1.03 liters of hydrogen.

Per Equation 2, one mole of hydrogen can create 2/3 mole of UH_3. Therefore, the water vapor could result in (0.038*2/3) ~ 0.026 gmoles of UH_3, or ~ 7 grams of UH_3.

Another source of hydrogen is radiolysis of the polyvinyl chloride bag containing the Pu-contaminated scrap. This has been previously evaluated (Reference 17) and the results of that evaluation is shown below.

Using conservative assumptions, it was calculated that uranium metal containing 1 gram of weapons-grade plutonium on its surface would generate hydrogen at a maximum rate of 70 cm^3/year. In nine years, it could produce 630 additional milliliters of hydrogen. This would generate about (630/(273*82.06)) ~ 0.026 gram-moles of hydrogen. If all of the hydrogen reacted to form UH_3, this would produce about 0.017 gram-moles of UH_3.

The total UH_3 that could form, using conservative assumptions for hydrogen production and then using the very conservative assumption that all hydrogen produced reacted with the uranium, forms only about (0.017+0.043) ~ 0.060 gram-moles of material. This is (0.060*238) ~ 14 grams of UH_3.

The density of uranium hydride is 10.91 grams/cc, and the density of uranium metal is 19 grams/cc (Reference 7). The density of uranium oxide is similar to the hydride. Because of the large decrease in density from uranium metal to its corrosion products (and associated increase in volume), only a thin film (< 750 micrometers) typically remains on the metal (Reference 7). Fourteen grams of UH_3 would occupy about two cm^3, if perfectly packed. In powder form, the pour density would be about 2 grams/cm^3,so fourteen grams of UH_3 powder would occupy about seven cm^3.

It is unlikely the hydrogen formed by radiolysis would form UH_3. This hydrogen would have to diffuse through the oxide layer to reach the metal surface. Chemisorption is slow, and splitting of the hydrogen molecule into hydrogen atoms on the oxide surface is endothermic and slow (Reference 7).

It is also unlikely that all the hydrogen formed will stay within the SLC, since it will diffuse through the vulcanized silicone rubber gasket. An exact solution of the rate requires solution of differential equations. A static calculation demonstrates the effect. This calculation is shown below.

The permeability of hydrogen through vulcanized silicone rubber is given in Reference 18. Permeability is a function of temperature, and is calculated from the following relationship, where all constant values are specifically for diffusion of hydrogen through vulcanized silicone rubber.

$$P = P_0*\exp(-E_p/(R*T)) \qquad \text{Equation 4}$$

Where:
P = permeability, cm^3-cm/(cm^2*sec*Pascal)
P_0 = pre-exponential factor, 0.1457E-07 cm^3-cm/(cm^2*sec*Pascal) (cm^3 at 0 °C, 1 atm)
Ep = Activation energy of permeation, 13.7 Kilojoules/mole
R = ideal gas constant, 8.3144E-03 Kilojoules/(mole-°K)
T = temperature, °K

Using a temperature of 20 °C (293 °K), the resulting permeability for hydrogen through vulcanized silicone rubber is 524E-11 cm^3-cm/(cm^2*sec*Pascal).

It is assumed the gasket is compressed from about 1/8-inch thickness to 1/16-inch (0.159 cm) thickness when the SLC is assembled. The diameter of the SLC is about 11.75-inches, so the circumference is (3.14159*11.75) ~ 36.9 inches. The thickness of the metal side of the SLC (where the gasket is compressed) is 1/16-inch (0.159 cm) thick. The area for diffusion is thus ((36.9/16)*6.54) ~ 15.09 cm^2. A driving force of 2% hydrogen (2026 Pascals) is assumed. There are 3.15E+07 seconds in a year. The diffusion of hydrogen per year from the PC, in cm^3 per year, becomes:

(5.24E-13*15.09*3.15E+07*2026/0.159) ~ 317 cm^3/year

At 273 °K and 1 atmosphere, this is (317/(273*82.06) ~ 0.014 gmoles/year. This is higher than the average production rate per year inside the inner bag, and demonstrates that diffusion from the SLC competes with reaction with metal for any hydrogen formed.

5.0 Assumptions

Any loose oxide was removed from the plutonium-contaminated scrap before it was shipped. This limits the amount of powdery material that would be in the bag when removed from the SLC.

The average temperature and relative humidity at SRS was assumed for the shipping origin.

There is a maximum of one gram of plutonium on the surface of any scrap piece.

6.0 Discussion

Because of geometry, the plutonium-contaminated scrap will not ignite during repackaging unless induced by UH_3. Using bounding assumptions, the amount of UH_3 that could be formed in up to nine years of storage in the POC is only 0.055 gram-moles. It is unlikely that the theoretical amount of hydride would form (Reference 19). Some UH_3 may be present in the corrosion film when the plutonium-contaminated scrap is unpacked, and some stabilization may occur, as evidenced by isolated "sparking" in the film. This is not a fire, and should be allowed to proceed to completion in the glovebox.

The likelihood of a fire during unpacking is at least less than the likelihood that the pipe component of the POC was improperly sealed, allowing much more water vapor to enter the container than in a properly-assembled container. Even if this happened, it is still not assured that sufficient UH_3 would be formed to present a fire hazard. The assembly of the pipe component was covered by procedure, and the procedure was implemented under a Conduct of Operations program. It is judged to be at least Extremely Unlikely that there will be a metal or UH_3 fire while repackaging the plutonium-contaminated scrap, although stabilization of minor amounts of hydride may occur at a higher frequency.

It is still considered prudent to be prepared for a metal or UH_3 fire, even though it is at least extremely unlikely one would occur and the consequences of a plutonium-contaminated scrap fire would be low. Preparations are discussed in more detail below.

7.0 Recommendations

Maintain a low inventory of combustibles in the glovebox used for removing the scrap from the plastic bags . Maintain a supply of magnesium oxide in this glovebox for use if needed. Maintain a metal container, and a cover for same, adequate to hold plutonium-contaminated scrap material if susceptibility for fire is suspected. Prepare contingency procedures and train personnel in actions to take if a fire occurs.

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Appendix A: Operations Steps for Repackaging Plutonium-Contaminated Scrap

APPENDIX A: NORMAL OPERATONS, REPACKAGING MIXED SCRAP

This list does not include radcon checks. It is anticipated that there will be radcon checks (smears, external field monitoring) each time a boundary is opened, or after transfer of material from a glovebox.

1. Receipt

The material will arrive in H-Area in a 12-inch pipe overpack. This incorporates a 55-gallon drum, with fiberboard packing material, surrounding a 12-inch pipe vessel with a flange on the top end. The 12-inch pipe will contain an inner vessel with a screw-lid can. The inner vessel will contain Pu-contaminated scrap, double-bagged. The inner bag is polyvinyl chloride, and the outer bag is polyethylene.

The receipt of material will follow normal receipt methods. The drum will be transported to as close to rooms 513 or 517 as practical, using a drum cart, before starting to unpack. MC&A/Safeguard and Security activities (to be specified later) will be performed on receipt.

2. Unpack and move scrap into hood

-Take top lid off drum, remove adequate fiberboard packing material to expose the 12-inch pipe flange.

-Take bolts out of pipe flange, remove flange

-Ensure combustible waste has been removed from hood/glove box before removing inner can from drum

-Using proper tool, take the inner can out of the pipe. This tool attaches to the top of the can when inserted into designed areas in the can top.

- Remove the empty drum from the hood area. Ensure the number of opened drums are consistent with facility combustible limits. This may be performed after the vessel is moved into the entry hood.

-Move inner vessel into entry hood

-Using proper tool, remove top cover from inner vessel

-Withdraw the bagged scrap from the inner vessel

-Pass the bagged scrap through the first passthrough port

-Pass the bagged scrap through the second passthrough port into the operating side of the process cabinet

-Place the bagged scrap in the pan supplied for scrap handling

-Inspect the bag for the presence of powdery material in the bottom of the bag

Actions if powdery material is observed are discussed later in this writeup. The following steps assume there was not a significant amount of powdery material in the bag.

-Cut through plastic bags (it is acceptable to cut through both bags in the same cut, if this is practical)

-Remove the scrap from the bags, then remove empty bags from immediate work vicinity (don't have to be bagged out at this point). Place in metal bin.

3. Repackaging scrap

-Perform required MC&A activities (to be specified later)

-Determine whether the scrap will pass through the go-no go gage (to determine required can size for repackaging)

-Collect any loose material remaining in the bag in the scrap handling, if there is any visible loose material in the bags

-Place loose material in small can or nylon bag and close

-Pass scrap and can (or nylon bag) through the bagout port into end of nylon bagout sleeve

-Ensure sufficient air has been expelled from the bagout sleeve containing the scrap so that the bag will not obstruct placing the scrap in the final can

-Heat-seal the scrap and small can (or small nylon bag) in the nylon bag formed from the bagout sleeve.

-Cut bagout sleeve to separate packaged scrap. Typically, three heat-sealed regions are formed. The bag is then cut through the middle of the middle seal to preserve glovebox integrity.

-Place the scrap into the dissolvable can, positioned to fit over post

-Seal lid to can by crimping or other suitable methods

-Place can on carrier

-Transfer can to H-Canyon using appropriate MC&A/Safeguards and Security protocols

Actions to Take For Reassembled Plutonium-Contaminated Scrap Parts

-There are a few bags (less than 15) that contain plutonium-contaminated scrap that was previously sampled, then reassembled for storage

-Unpack plutonium-contaminated scrap from bags as instructed above, with or without the presence of significant powdery material

-Remove samples from primary scrap material

-Dispose of samples in a manner consistent with criticality restrictions

Actions to Take If Significant (Nominally >2 cm^3) Of Powdery Material in Bottom of Bag

(Note: It is considered extremely unlikely that significant powdery material will be in the bags. X-Rays taken to date in KAMS of a representative sample of the containers confirms no powdery material has collected to date. However, the following steps are included in case this contingency occurs.)

-Carefully remove outer polyethylene bag and remove from vicinity of scrap

-Reposition metal container previously installed in glovebox for this use

-Carefully place the inner bag into the container

-Carefully cut a small hole (nominally one square centimeter) in the top of the PVC bag to allow air to diffuse into the bag. The goal is to introduce oxygen slowly into the bag, allowing any UH_3 to react slowly.

-Allow sufficient time (to be determined later) for oxygen to stabilize any UH_3 present in the bag

-If flames are seen, add magnesium oxide sand as appropriate and place cover on lid to limit rate of combustion

-After stabilization, remove the plutonium-contaminated scrap from the bag and continue with normal operations for the material

-Empty powdery material remaining in the bag into a tray, allow to continue stabilizing if needed

-Sweep the powder and place in a small can or bag; package with the scrap material in the dissolvable can