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**DEFENSE NUCLEAR FACILITIES
SAFETY BOARD**

Washington, DC 20004-2901



January 3, 2013

Mr. David Huizenga
Senior Advisor for Environmental
Management
U.S. Department of Energy
1000 Independence Avenue, SW
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Dear Mr. Huizenga:

The Department of Energy (DOE) stores reactive uranium and thorium metal spent nuclear fuel (SNF) underwater at the L-Reactor Disassembly Basin (L-Basin) at the Savannah River Site (SRS). The Board is encouraged that DOE has begun processing some of this fuel, the Sodium Reactor Experiment fuel, in H-Canyon at SRS. This action will eliminate the vulnerability of long-term, underwater storage of this type of reactive metal SNF. However, further attention to the disposition of other vulnerable fuel types remaining in the L-Basin is warranted. Enclosed is DNFSB/TECH-38, *Storage Conditions of Reactive Metal Fuel in L-Basin at the Savannah River Site*. This report, which is provided for your information, identifies the vulnerabilities associated with interim and long-term storage of SNF in L-Basin.

Sincerely,

Peter S. Winokur, Ph.D.
Chairman

Enclosure

c: Mr. David C. Moody, III
Mrs. Mari-Jo Campagnone

Storage Conditions of Reactive Metal Fuel in L-Basin at the Savannah River Site

Defense Nuclear Facilities Safety Board Technical Report



January 2013

Storage Conditions of Reactive Metal Fuel in L-Basin at the Savannah River Site



This technical report was prepared for the Defense Nuclear Facilities Safety Board by staff members:

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EXECUTIVE SUMMARY

A portion of the spent nuclear fuel (SNF) inventory of L-Reactor Disassembly Basin (L-Basin) at the Savannah River Site (SRS) consists of uranium, thorium, and other similarly reactive metal SNF assemblies from various test and experimental reactors. Water can react with this metal fuel to form an oxide and release hydrogen gas. Oxide formation can cause handling problems due to sludge formation, as well as rupture of the storage cans due to the accompanying volume increase and the release of soluble fission products. The hydrogen gas can accumulate or lead to the formation of hydrides that can spontaneously ignite in air. Much of the reactive metal SNF inventory in L-Basin is vulnerable to these metal-water reactions because one or more of the following conditions are present:

- The cladding has ruptured, is missing, or was breached when the fuel assembly was cut into pieces, thus allowing water, if present, to contact the metal fuel directly.
- The metal fuel was initially packaged in direct contact with water.
- The fuel was initially packaged dry inside a sealed can, but the can is known to have failed during storage, allowing water to enter the can.
- The fuel is stored inside cans whose designs are vulnerable to developing leaks.
- The cans are heavily corroded as a result of past storage conditions and are still vulnerable to ongoing pit corrosion.

The limited inspection data indicate that many of the cans have significant corrosion and that some have failed leading to fuel degradation. Gas is being released periodically from several of the cans, indicating that the metal fuel is continuing to degrade. As the fuel degrades it becomes more difficult to handle, repackage, and/or process in the future. The Department of Energy currently is not equipped to handle or repackage degraded fuel in L-Basin. Further attention to the disposition of vulnerable fuel types that remain in the L-Basin is warranted.

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1. BACKGROUND

The Savannah River Site (SRS) currently stores spent nuclear fuel (SNF) underwater in the L-Reactor Disassembly Basin (L-Basin). The majority of the fuel is now from foreign and domestic research reactors, but in the past it was dominated by defense fuel (Mk16 and Mk22) from SRS's production reactors. In 1994, the Defense Nuclear Facilities Safety Board (Board) issued Recommendation 94-1, *Improved Schedule for Remediation in the Defense Nuclear Facilities Complex* (Defense Nuclear Facilities Safety Board, 1994). Subrecommendation 6 recommended:

That preparations be expedited to process the deteriorating irradiated reactor fuel stored in basins at the Savannah River Site into a form suitable for safe interim storage until an option for ultimate disposition is selected.

The Department of Energy's (DOE) response to this Subrecommendation (Department of Energy, 1994) entailed the dissolution of Mk16/Mk22 aluminum-clad SNF at SRS and the completion of several upgrades to L-Basin to improve water chemistry and reduce the corrosion rate of the remaining SNF.

In addition to aluminum-clad SNF, SRS has a smaller inventory of zirconium alloy-clad SNF. In the *Record of Decision (ROD) on Programmatic Spent Nuclear Fuel Management and Idaho National Engineering Laboratory¹ Environmental Restoration and Waste Management Programs* (U. S. Department of Energy, 1995), DOE promulgated its decision that "non-aluminum-clad fuels...would be transferred to the Idaho National Engineering Laboratory." At that time, DOE noted that the "No Action" alternative "would adversely affect the Department's mission to ensure safe and secure management of spent nuclear fuel. Future deterioration of fuels and facilities may increase accident risks." The *Record of Decision for the Savannah River Site Spent Nuclear Fuel Management Environmental Impact Statement, Aiken, SC* (U.S. Department of Energy, 2000) reiterated this decision,² along with the plan to use conventional processing to stabilize the Sodium Reactor Experiment (SRE) SNF. The exchange of SNF with Idaho National Laboratory (INL) did not happen. The SRE SNF processing campaign in H-Canyon commenced during August 2012.

With the exception of the SRE SNF, the SNF inventory in L-Basin lacks a disposition pathway. While this situation is not ideal for the aluminum-clad research reactor fuel, its behavior in wet storage is fairly well understood, and the technologies for processing it have been demonstrated and are currently available in H-Canyon. On the other hand, the zirconium alloy-clad SNF is not well characterized and there is evidence that some of it is degraded. This fuel was not processed at SRS in the past. While it can be processed, new mechanical operations would be required to fully expose the metal fuel to the dissolution chemicals used in H-Canyon (Defense Nuclear Facilities Safety Board, 1999).

¹ Idaho National Engineering Laboratory was also known as Idaho National Laboratory and Idaho National Engineering and Environmental Laboratory when some of the references cited herein were published.

² Although not directly stated in the ROD, DOE planned to store this SNF in dry storage at INL pending final disposition decisions.

2. FUEL DEGRADATION

The fuels that are most likely to degrade (or degrade further) during long-term storage are uranium and thorium metal fuels that are not protected from water with a metallurgically bound cladding. At L-Basin, the uranium and thorium metal SNF inventory consists largely of Heavy Water Components Test Reactor (HWCTR) fuel assemblies. These assemblies normally contain natural uranium, although some were fabricated with low or highly enriched uranium. Most of these items consist of uranium metal fuel with zirconium alloy cladding. In some cases, the uranium fuel is alloyed with small amounts (less than 2 percent) of zirconium, molybdenum, or silicon. These alloying elements do not significantly change the metal's reactivity with water. A few items are predominantly thorium metal. For the purposes of this report, "metal" refers to all of these items that react similarly to uranium metal in water.

Water reacts with uranium metal to form uranium oxide. Since uranium oxide is less dense than uranium metal, this reaction creates a volume increase that can lead to ruptured cladding or a breached container. Well-oxygenated water can form adherent oxide layers that protect the remaining fuel from further degradation. If the water in contact with the fuel becomes anoxic, however, uranium oxide formed by the corrosion of the uranium metal can begin to spall instead of remaining as a protective layer. Thus, the uranium metal corrosion rate may be 10 to 1,000 times greater than that caused by aerated water (Pacific Northwest National Laboratory, 2008). Although the majority of the HWCTR fuel has a zirconium alloy cladding, DOE cut most of the fuel assemblies into pieces, and the cladding sometimes ruptured during irradiation. Once water comes in contact with the exposed metal fuel, the expansion resulting from the corrosion of the fuel can cause splitting of the remaining cladding, exposing more metal fuel and accelerating the degradation process. The end result of this degradation is often a loose sludge that can be difficult to recover and stabilize.

In addition, hydrogen can be generated by either the corrosion of uranium metal or the radiolysis of water. When water leaks into a breached can that contains metal fuel, the hydrogen formed through corrosion or radiolysis can escape from the can or accumulate in the headspace, depending on the location of the breach in the can. If hydrogen gas accumulates, it can reach a flammable concentration in the can's headspace. The hydrogen formed as a result of corrosion or radiolysis can also react with uranium metal to form pyrophoric uranium hydride. This uranium hydride can react with water to form uranium oxide, freeing up more hydrogen to react with additional metal or escape into the water or gas space.

3. STORAGE CONDITIONS

3.1 Past Storage Conditions

Metal spent fuel at SRS is stored in a wide variety of can types. These cans are usually made of aluminum. The cans may be sealed, open, or have a J tube.

- Sealed cans keep the fuel inside dry and prevent water from corroding the reactive metal fuel as long as the seal remains intact. Sealed cans include K and Z cans, welded cans, and those equipped with a screw-top lid.
- Open cans have holes that allow water to enter the can or bundle and contact the fuel. If reactive metal fuel is exposed, the water may corrode the fuel and allow soluble fission products to leach into the water. Because the water inside and outside the can are allowed to mix, the soluble fission products may later leave the can. Depending on the location of the hole(s), insoluble corrosion products and any gases generated inside the can may be able to escape. Open cans include slug cans, failed element containers (FEC), and L bundles.
- A J tube can has a tube on the top of the can that is bent into the shape of an inverted J or U. The bend is filled with gas while the can itself is filled with water. The tube is open ended to allow any gas generated inside the can to escape. The gas pocket inside the tube prevents water inside the can from mixing with water outside the can. Since J tube cans are filled with water, water may be able to react with any exposed reactive metal fuel. Any soluble fission products that leach out of the fuel or insoluble corrosion products will remain inside the J tube can since the water inside the can is isolated from the surrounding water. J tube cans may be small inner cans or larger outer cans like the HWCTR and L oversized cans. Figure 1 illustrates an example of a J tube can design.

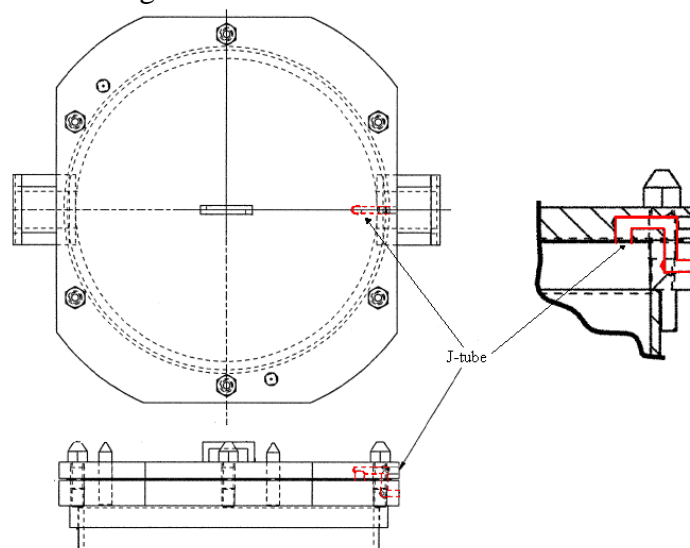


Figure 1. A J tube can design.

Various combinations of smaller cans are currently nested in L oversized cans or L bundles. The inner cans may be sealed, open, or have a J tube. Three of the more common inner can designs include K, Z, and slug cans.

- K can—A 9-inch-long aluminum can with a mechanical seal. A plate on top of the can is held in place with an aluminum bar. The top plate has a metal-to-metal seal with the can body, and the bar is wedged on top of the plate to hold the plate down and keep water out. See Figure 2.
- Slug can—A small can used to store fuel slugs. All slug cans have a small hole in the lid that allows water to enter.
- Z can—A long aluminum can that uses a plate and bolts to form a mechanical seal at the top. See Figure 3.



Figure 2. A new K can sitting on a drum next to a ruler.



Figure 3. Lower portion of a cracked Z can.

Small inner cans like K and slug cans may be stacked inside a FEC. The FEC is a 5-inch-diameter, 12-foot-long aluminum can that has an open vent tube on top. The FEC is then stored inside a HWCTR or L oversized can. See Figure 4.

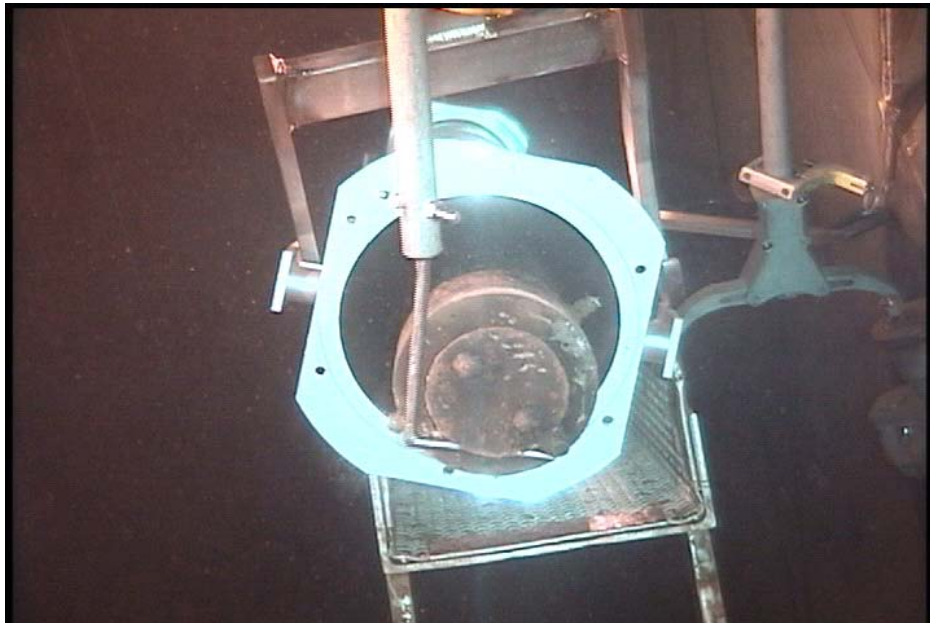


Figure 4. A failed element container being loaded into an L oversized can.

In the past, inner cans and FECs were stored inside buckets or a HWCTR oversized can. Today, inner cans and FECs are stored inside a L oversized can or a L bundle.

- HWCTR oversized can—A 14-inch-diameter, 14-foot-tall older outer can made of either aluminum or stainless steel, equipped with connections for sampling and flushing, that is no longer used. See Figure 5.
- L oversized can—Currently used as an outer can, it is similar to the HWCTR oversized can but only 8 inches in diameter. These aluminum cans have J tubes, but no connections for sampling or flushing. See Figure 6.
- L bundle—A 5-inch-diameter, approximately 11-foot-long aluminum tube currently used as an outer can. The bottom plate and lid each have eight 0.25-inch diameter holes that allow the basin water to enter. See Figure 7.

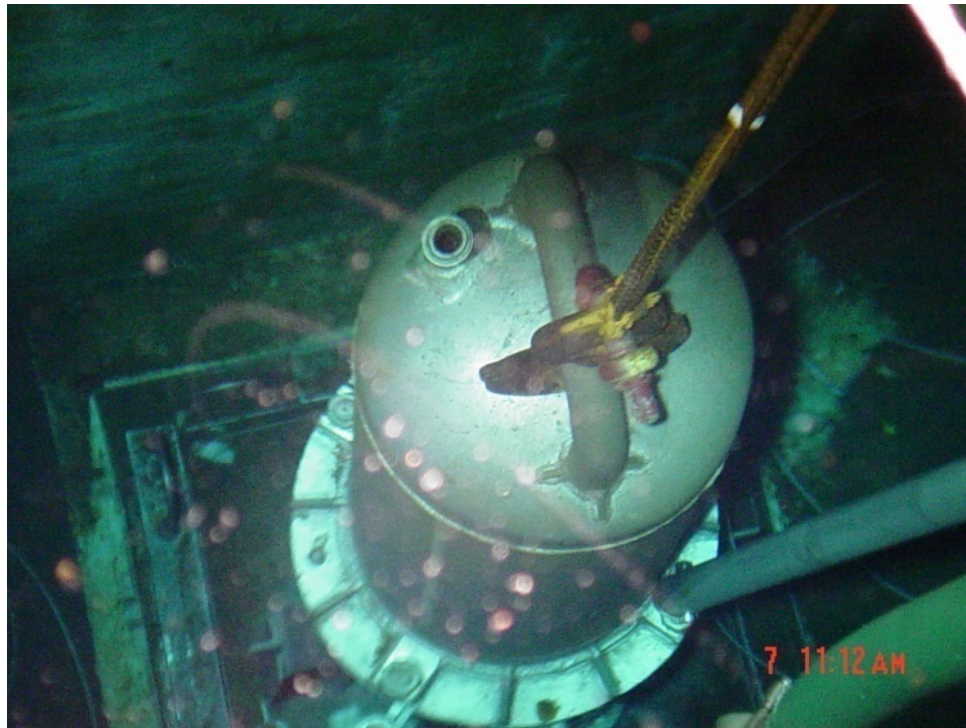


Figure 5. An aluminum HWCTR oversized can



Figure 6. Installing the lid on an L oversized can.

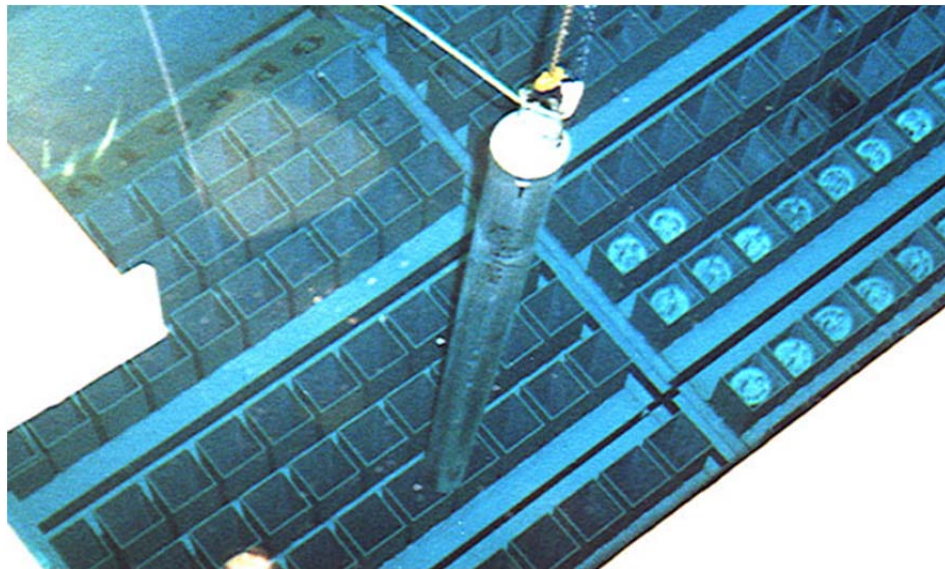


Figure 7: An L bundle being raised above storage racks.

DOE irradiated the HWCTR test assemblies in R-Reactor between 1957 and 1963 as part of the Power Reactor Program. After examination and destructive testing, DOE placed the fuel pieces in aluminum cans (e.g., K and Z cans, depending on their size) and stored them in R-Basin. These cans usually had a seal to prevent the fuel from contacting the basin water. When R-Reactor shut down in 1964, DOE moved these assemblies to P-Basin for the remainder of the 1960s. At P-Basin, DOE suspected that some of the smaller sealed cans were leaking (reportedly

because bubbles were being released) and placed them inside FECs. In other cases, DOE packaged cut-up or intact fuel assemblies in direct contact with water inside a can equipped with a J tube. During 1969 and 1970, DOE moved the HWCTR fuel assemblies to the Receiving Basin for Off-Site Fuel (RBOF), where many of the cans were stored inside buckets. DOE repacked several of the sealed cans, FECs, and intact fuel assemblies into nine HWCTR oversized cans. These HWCTR oversized cans were designed to store failed fuel and contained J tubes. Although the HWCTR oversized cans had connections for sampling and flushing, DOE did not open or flush them for nearly 30 years, although they were known to contain leaking containers because gas bubbles could be observed venting through the J tubes. In 1980, DOE consolidated the contents of many of the remaining small sealed cans into several stainless steel cans with J tubes.

After approximately 30 years of storage at RBOF, DOE deinventoried RBOF between 1997 and 2003 and moved all of the HWCTR fuel assemblies to L-Basin. As part of this effort, DOE repacked the fuel assemblies from the original HWCTR oversized cans into smaller L oversized cans. DOE flushed several HWCTR oversized cans to remove the radionuclides that had accumulated in the water within the cans as a result of degradation of the SNF. These HWCTR oversized cans were opened, at which time it was noted that half of the Z cans inside were cracked (see Figure 8). The cracks allowed water to enter the cans and react with the uranium metal fuel to form uranium oxide and possibly uranium hydride. In one case, the corrosion of the metal fuel resulted in the deposition of 36 pounds of corrosion products at the bottom of just one HWCTR oversized can. In addition, the water leached out soluble fission products such as cesium-137 (Cs-137). Therefore, DOE decided to flush several of the HWCTR oversized cans using an underwater pump, pre-filter, and resin deionizer system. This process captured 319 curies of leached Cs-137 in the resin and an additional 12 pounds of corrosion product fines in the filter. Although DOE placed the cracked cans inside new L oversized cans, the damaged fuel remains in contact with water, where it can continue to degrade. DOE also packaged the pre-filters, the ion exchange resin, and the corrosion products at the bottom of the worst HWCTR oversized can into cans with J tubes and shipped them to L-Basin because it had no way to handle them. DOE loaded all of the other J tube cans in RBOF and the small sealed cans in the buckets into L bundles.



Figure 8. Cracked Z can inside an HWCTR oversized can.

3.2 L-Basin Storage Today

HWCTR and other metal fuel is stored in L oversized cans, L bundles, and other miscellaneous containers underwater in L-Basin. In many cases, the fuel assemblies are in direct contact with water because the inner can was originally filled with water, a weld or seal failed, or the can cracked. A summary of key items in the L oversized can inventory is as follows:

- Seven L oversized cans contain a total of 13 cans with screw-top lids, 52 slug cans, and 71 K cans. In many cases, these smaller cans are nested inside 12-foot-long, vented FECs that sit inside the L oversized can. The slug cans have a hole drilled in the lid that allows water to enter and contact the fuel slugs. The K cans are sealed by a bar wedged on the top plate to form a metal-to-metal seal. In a 2005 report, a Westinghouse Savannah River Company engineer concluded that some of the “seals have likely failed after 40 years of underwater storage” (Westinghouse Savannah River Company, 2005). Bubbles are being released from the vents of these seven L oversized cans, including those that contain only sealed K cans. This suggests that some of the seals of the approximately 50-year-old K cans have failed. Furthermore, when DOE flushed the HWCTR oversized cans that contained these FECs, the dose rates measured on the exterior of the HWCTR oversized cans sometimes dropped 300–400 mrem/hr, which indicates that fission products (e.g., Cs-137) had leached out of the fuel inside the slug and/or K cans and were carried away during the flush process.

- Four L oversized cans contain a total of six Z cans, including the three that previously failed. The Z cans have a plate wedged in on top that is held in place with a bolt. Bubbles are being released from the vents of these four L oversized cans, including those that contain only Z cans that were still intact in 2002. This indicates that the fuel in the breached Z cans is still reacting with water and that additional Z cans have failed since they were initially repacked.
- Two L oversized cans contain intact fuel assemblies. The assembly in one L oversized can is in direct contact with water, while the other assembly is inside a sealed can. An inspection found no bubbles being released during the observation period. This could mean that the cladding or can seal may still be protecting the uranium metal or that the reaction is slow or has stopped.

DOE stores metal fuel in about two dozen L bundles. In some cases, a special plate was added to the bottom of the L bundle to prevent any corrosion products from falling out of the L bundle. A summary of key items in the L bundle inventory is as follows:

- Several L bundles contain one or more K cans.
- Seven L bundles each contain a fuel assembly inside a sealed aluminum tube with a J tube vent in the lid. Some of the assemblies are intact, while others have ruptured cladding. The metal fuel in each of the tubes is in contact with water. During the RBOF deinventory campaign, DOE found that one of the tubes had failed (see Figure 9). All of these items were placed in L bundles that were modified to include a solid bottom to contain corrosion products and a zeolite filter in the lid to capture cesium that was released.
- Six L bundles contain intact uranium and thorium metal fuel assemblies in direct contact with water. The 50-year-old cladding on the fuel assemblies is the only barrier protecting the metal.
- The remaining L bundles contain fuel assemblies inside cans, some with and some without J tubes. Each of these inner cans is filled with water.



Figure 9. Sealed tube with crack at bottom.

L-Basin also contains other water-reactive fuels inside L bundles. Thirty-seven L bundles contain thorium-uranium metal and uranium carbide fuel from the SRE (Westinghouse Savannah River Company, 2005). These fuels are stored inside welded shipping cans. While the fuel assemblies are intact, they no longer have any cladding, so the fuel would readily react if water leaked inside the cans. An additional four L bundles contain destructively tested and disassembled fuel plates from the Experimental Boiling Water Reactor. The water-reactive fuel is stored inside welded cans to keep water out. One of the can welds failed in 1999. Later, DOE put the breached can inside a L bundle and shipped it to L Basin.

3.3 Can Corrosion

Nearly all the inner cans containing metal fuel are approximately 50 years old, and DOE is considering the possibility of extended storage of these cans for an additional 50 years (U.S. Department of Energy Savannah River Operations Office, 2011a). Many of these cans were stored in P-Basin and R-Basin for 6 to 12 years (see Figure 10). The water in these basins was untreated, filtered river water. Even though the L oversized cans and L bundles are now stored in L-Basin, which is filled with treated water, the water inside the L oversized cans and the smaller J tube cans inside L bundles is isolated from the basin water. Thus, the water contained in the smaller J tube cans stored in L bundles is the water present in P-Basin and RBOF when the cans were loaded in the 1960s and 1980, while the water in the L oversized cans is from RBOF 10 years ago. Although the L bundles have holes in the lids and some bottom plates to allow basin water to enter, the amount of mixing is limited by the $\frac{1}{4}$ -inch size of the holes; the long, narrow shape of the tube; the obstructions caused by the inner cans or fuel assemblies; and the relatively low decay heat of the fuel, which would not create much of a thermal gradient to drive convection. The current designs of the L oversized cans and the smaller J tube cans do not allow the water inside to be sampled or flushed.

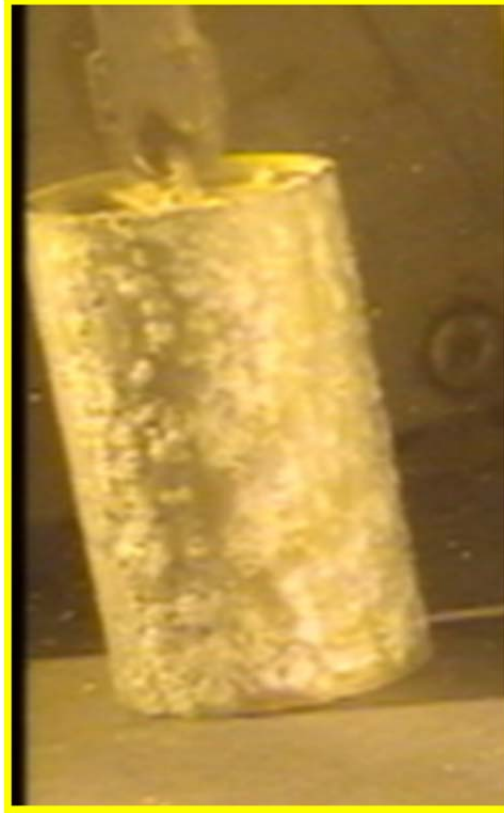


Figure 10. Aluminum can previously exposed to P-Basin water.

DOE has very limited data on the condition of the various cans in use. During the RBOF deinventory campaign a decade ago, DOE was able to inspect the exterior of the small cans in the RBOF buckets along with two small cans, FECs, and Z cans inside the HWCTR oversized cans. DOE noted substantial surface pitting (see Figures 11 to 13). Once pitting has begun on the surface of a storage container, the local electrochemical conditions within the pit, not the bulk chemical composition of the fluid outside the pit, drive continued pit penetration into the metal. Once initiated in the storage cans, pits can continue to propagate regardless of the purity of the water within the L oversized can or L bundle.



Figure 11. K can with evidence of pitting from one of the HWCTR oversized cans that did not require flushing.



Note: This is not one of the three cracked Z cans.

Figure 12. Z can with evidence of pitting removed from an HWCTR oversized can.



Figure 13. Failed element container with evidence of pitting.

4. Current Situation

At the Board's June 16, 2011, public meeting in Augusta, Georgia, the Board noted that some of the SNF items at SRS were not good candidates for long-term storage—particularly the cans of SRE clad thorium/uranium metal SNF that could react with water if the cans leaked. Further, the Board questioned the indefinite storage of damaged and cut SNF because the current condition of many of these items remains unknown, DOE has not inspected them since they were packaged decades ago, and most of the inner cans are already 50 years old. The approach chosen by DOE in response to the Board's Recommendation 94-1, *Improved Schedule for Remediation in the Defense Nuclear Facilities Complex* (U.S. Department of Energy, 1994), is a good example of the types of actions that would benefit this situation. To support that Recommendation, DOE conducted a complete underwater video inspection of all SNF and storage equipment at INL and nondestructively examined canisters to determine the condition of the canisters and the SNF they contained.

On November 22, 2011, DOE concluded that the SRE SNF, which has no cladding any more, was "more vulnerable to long-term wet storage due to the metallic composition of the fuel and its reactivity with water if a can breach occurs" (U.S. Department of Energy Savannah River Operations Office, 2011b). As a result, DOE directed its contractor to immediately initiate the required preparations for processing the SRE SNF in H-Canyon. The SRE SNF processing campaign began in August 2012.

As described in this report, however, the storage conditions for the rest of the metal SNF in L-Basin are not as robust as those DOE implemented for similar deteriorating metal SNF in response to Recommendation 94-1. Further attention to the disposition of the other vulnerable fuel types remaining in the L-Basin is warranted.

REFERENCES

Defense Nuclear Facilities Safety Board, 1994, *Improved Schedule for Remediation in the Defense Nuclear Facilities Complex*, Recommendation 94-1, May 26.

Defense Nuclear Facilities Safety Board, 1999, *Savannah River Site Spent Nuclear Fuel*, DNFSB/TECH-22, April 21.

Pacific Northwest National Laboratory, 2008, *Uranium Metal Reaction Behavior in Water, Sludge, and Grout Matrices*, PNNL-17815, September.

U.S. Department of Energy, 1994, *Defense Nuclear Facilities Safety Board Recommendation 94-1 Implementation Plan*, December 5.

U.S. Department of Energy, 1995, *Record of Decision on Programmatic Spent Nuclear Fuel Management and Idaho National Engineering Laboratory Environmental Restoration and Waste Management Programs*, 60 FR 28680, June 1.

U.S. Department of Energy, 2000, *Record of Decision for the Savannah River Site Spent Nuclear Fuel Management Environmental Impact Statement*, Aiken, SC, 65 FR 48224, August 7.

U.S. Department of Energy Savannah River Operations Office, 2011a, *Direction Regarding an Augmented Surveillance & Maintenance Plan for L-Area and Resumption Planning for H-Canyon*, NMPD-11-0048, Letter from Patrick W. McGuire to Paul Hunt, June 30.

U.S. Department of Energy Savannah River Operations Office, 2011b, *Direction on Preparation for Processing the Initial Vulnerable Used Nuclear Fuels*, AMNMSP-12-0004, Letter from Charlene Smith to John Temple, November 22.

Westinghouse Savannah River Company, 2005, *Spent Fuel Isolation Cans Inventory*, M-TRT-L-0003, March.

