DEFENSE NUCLEAR FACILITIES SAFETY BOARD

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November 9, 1995

The Honorable Thomas P. Grumbly
Assistant Secretary for Environmental Management
Department of Energy
Washington, DC 20585-0113

Dear Mr. Grumbly:

Please find enclosed for your use a report entitled, *Stabilization of Deteriorating Mark 16 and Mark 22 Aluminum-Alloy Spent Nuclear Fuel at the Savannah River Site* (DNFSB/TECH-7). This report, prepared by a Defense Nuclear Facilities Safety Board (Board) staff team, examines the dry storage and chemical processing alternatives for stabilizing the Mark 16 and Mark 22 spent fuel. The report concludes that stabilization of the spent fuel by chemical separation, as originally proposed by the Department of Energy in Response to Board Recommendation 94-1, is the best alternative for meeting the intent of Recommendation 94-1.

Sincerely,

John T. Conway
Chairman

Enclosure

c: The Honorable Charles B. Curtis
    The Honorable Tara J. O'Toole
    The Honorable Victor H. Reis
    Mr. Mark Whitaker

STABILIZATION OF DETERIORATING MARK 16 AND MARK 22 ALUMINUM-ALLOY SPENT NUCLEAR FUEL AT THE SAVANNAH RIVER SITE

Defense Nuclear Facilities Safety Board

Technical Report

November 1, 1995

STABILIZATION OF DETERIORATING
MARK 16 AND MARK 22
ALUMINUM-ALLOY SPENT NUCLEAR FUEL
AT THE SAVANNAH RIVER SITE

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I. EXECUTIVE SUMMARY

A significant portion of the Department of Energy (DOE) spent nuclear fuel at the Savannah River Site (SRS) is deteriorating due to corrosion. Specifically, the Mark 16 and Mark 22 spent fuel have been improperly stored and have experienced corrosion and degradation of the aluminum cladding and the aluminum-uranium core.

DOE is pursuing a strategy to dispose of DOE-owned spent fuel in a geologic repository by the year 2035.[1] However, wet-stored fuel has suffered corrosion, releasing fission products into the storage water. This deteriorating spent fuel needs to be stabilized to arrest further deterioration and to allow safe interim storage.

In the Defense Nuclear Facilities Safety Board (Board) Recommendation 94-1, the Board expressed special interest in "processing canyons and reactor basins at the Savannah River Site contain[ing] large amounts of deteriorating irradiated reactor fuel ...."[2] The Board stated its concern about the slow pace of remediation and recommended "that preparations be expedited to process the deteriorating irradiated reactor fuel ... into a form suitable for safe interim storage ...." DOE accepted this recommendation.[3]

DOE's Implementation Plan for Board Recommendation 94-1 proposed chemical processing to stabilize the SRS fuel.[4] Currently, DOE is reevaluating two methods of stabilizing the deteriorating Mark 16 and Mark 22 aluminum-alloy spent fuel: chemical processing to a low-enriched oxide and dry storage. A comparison between these methods is presented in this report, and the conclusion is summarized below.

Stabilization of the spent fuel by chemical processing:

- Reduces the risk associated with continued wet storage and subsequent dry storage of the deteriorating fuel.

- Eliminates the need for additional processing in the future that would involve additional safety risks.

- Produces the smallest volume of high-level waste.

- Provides the lowest cost alternative.

This report concludes that stabilization by chemical separation, originally proposed by DOE in response to Board Recommendation 94-1, is the best alternative for remediating the Mark 16 and Mark 22 spent fuel.

II.   INTRODUCTION / BACKGROUND

For about 30 years, heavy-water reactors at the SRS were operated for defense-production of nuclear materials. Low-temperature reactor operation allowed the use of aluminum-clad, aluminum-alloy fuel, such as the Mark 16/22 fuel. This design facilitated both fuel fabrication and subsequent spent fuel reprocessing. At the end of a reactor cycle, the Mark 16/22 fuel was normally discharged to cooling basins and stored for up to eighteen months. After cooling, the fuel was chemically processed in H-Canyon to recover the highly enriched uranium.

In April 1992, with chemical separations activities already temporarily suspended, DOE implemented a decision to phase out defense-related chemical separation at the SRS. Processing of the "in process" irradiated nuclear materials, including spent fuel, was not completed. This course of action left irradiated material in interim forms and locations that were not designed for long-term storage.

Recommendation 94-1: In May 1994, the Board issued Recommendation 94-1.[2] In this recommendation the Board expressed special interest in "... processing canyons and reactor basins at the Savannah River Site contain[ing] large amounts of deteriorating irradiated reactor fuel ...." The Board observed that "... the halt in production of nuclear weapons and materials to be used in nuclear weapons froze the manufacturing pipeline in a state that, for safety reasons, should not be allowed to persist unremediated." The Board stated its concern about the slow pace of remediation and 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."

DOE accepted this recommendation on August 31, 1994,[3] and issued an implementation plan in February 1995.[4] In this implementation plan, DOE stated:

> "The vast majority of DOE-owned spent nuclear fuel was designed to be reprocessed and is therefore susceptible to dissolution in aqueous solutions. Long-term storage in the underwater fuel storage facilities was not intended for the majority of the spent fuel. The storage facility engineering design and the monitoring requirements were not adequate to compensate for the various underwater corrosion mechanisms experienced due to the extended storage. Severe unintended consequences have resulted, including the loss of configuration control of the storage equipment; the failure of cladding, which affects criticality safety, sludge generation, and fuel handling; and radionuclide leakage into the basin water, which affects personnel exposure and increases potential environmental impacts."

The implementation plan went on to say:

> "Because these facilities were designed between 30 and 50 years ago, most do not meet all current standards for seismic resistance to prevent potential fuel reconfiguration or current standards for leak protection and detection. A design basis seismic event may result in reconfiguration of fissile material and potential criticality, worker overexposure, and leakage to the environment. Inaccurate leak detection and [in]adequate barriers to leakage could result in unmonitored releases of radioactive material to the environment."

DOE agreed that "... the materials addressed by the Board should be converted into a form suitable for safe interim storage on a high-priority basis." For the SRS, the implementation plan set objectives to:

- Complete dissolution of the Mark 31 targets* by September 1996.

- Complete dissolution of Mark 16/22 spent fuel by November 1999.

- Complete processing of the resultant uranium solutions by April 2000.

In addition to the objective of converting the deteriorating spent fuel into a form suitable for safe storage, DOE identified actions to limit the extent of fuel corrosion during continued wet storage prior to processing. Most of the actions identified were to improve water quality and included shock-deionization, installation of continuous deionization systems, installation of deionized makeup water systems, and removal of basin sludge. Another action identified to limit fuel corrosion was the relocation of vertically stored fuel to horizontal storage racks (to eliminate a stainless steel to aluminum galvanic couple). All of these actions are to be completed by September 1997.[4]

NEPA Considerations: In its 94-1 Implementation Plan, DOE also stated its commitment "to a comprehensive National Environmental Policy Act (NEPA) review process in making decisions on the storage, disposition, and, if appropriate, transportation of DOE-owned spent nuclear fuel." To this end, DOE prepared an Environmental Impact Statement (EIS), entitled "Interim Management of Nuclear Materials (IMNM) at the Savannah River Site."[5] The purpose of this EIS was to determine what materials were stable and could safely remain in their current form for an interim period (approximately 10 years), and what materials were at risk and therefore required near-term stabilization to assure continued safe management. The EIS also addressed stabilization methods that might be used to achieve near-term stabilization.

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* The Mark 31 targets are aluminum-clad, depleted uranium targets used in the production of plutonium. A Mark 31 target assembly consists of 18 Mark 31 slugs.

The draft IMNM EIS was issued in March 1995. Quoting the Spent Fuel Working Group Report,[6] the draft IMNM EIS noted that:

> "Corrosion of fuel and target materials in the water basins and its effects constitute the major ES&H (Environment, Safety and Health) vulnerability at the SRS pertaining to stored Reactor Irradiated Nuclear Material. Corrosion is occurring in K- and L-Reactor basins and it is becoming increasingly difficult to maintain the cesium-137 activity within the administrative limit. Continued corrosion will eventually impact the physical integrity of stored materials. Such an eventuality would impact criticality, personnel radiation exposure, and fuel retrievability and disposal. The mechanisms and consequences of the corrosion are being addressed by WSRC (Westinghouse Savannah River Company) and the levels of contamination are low, however, fissile material such as uranium [and] plutonium are being released to the basin water which constitutes an ES&H vulnerability. Left unmitigated, the long term consequences of this situation could be severe."

Having categorized the Mark 16/22 fuel as requiring near-term stabilization, the draft IMNM EIS identified the preferred method for achieving this stabilization to be "Blending Down to Low-Enriched Uranium." This alternative would utilize chemical separation to convert the material into a form more suitable for long-term storage. Specifically, the highly enriched fuel would be dissolved in the H-Canyon. The fission products would be transferred to the high-level waste tanks for subsequent vitrification. The enriched uranium solution would be blended with depleted uranium and converted to a stable low-enriched uranium oxide.

Reconsidering Methods of Stabilization: In October 1995, the final IMNM EIS was issued and changed the preferred alternative for the Mark 16/22 fuels to the "No Action" alternative. The "No Action" alternative would result in continued storage of fuel in water-filled basins. This alternative postpones the selection of an appropriate stabilization method for the Mark 16/22 spent fuel. Postponing this selection allowed DOE to finalize the IMNM EIS. DOE is currently comparing dry storage and chemical processing in preparation for selecting the method of stabilization for the Mark 16/22 spent fuel.

III. CURRENT STORAGE CONDITION

A. Inventory

The SRS defense-production spent fuel has been in wet storage since 1988.** The Mark 16/22 spent fuel is currently being stored in several facilities, including the Reactor Disassembly Basins (K-Basin, L-Basin and P-Basin), the H-Canyon Storage Basin, and the Receiving Basin for Offsite Fuel (RBOF). Table 1 summarizes the inventories, as reported in the DOE Spent Fuel Working Group Report in November 1993.[6]

Facility	Mark 22	Mark 16	Mark 31
K-Basin	900	1	3
L-Basin	-	517	347
P-Basin	396	53	-
RBOF	-	2	-
H-Canyon	-	13	-
F-Canyon	-	-	68
Total	1296	586	418

Table 1: Inventory of SRS Defense-Production Irradiated Fuel and Target Assemblies (Mark 16/22/31). For the Mark 31, one assembly contains 18 slugs.

DOE has already concluded that the corroded Mark 31 targets should be processed.[5] Table 2 compares the Mark 31 heavy metal mass and fission product activity to the Mark 16/22 spent fuel. This table indicates that, while the Mark 31 targets have 23 times the mass of heavy metal, the Mark 16/22 spent fuel has 23 times the fission product inventory. This fission product inventory constitutes a significant hazard and is dominated by radionuclides with approximately 30 year half-lives, assuming a decay time of seven years (i.e., 85 percent strontium-90, cesium-137, and their daughters). To put the fission product inventory in perspective, if one percent of the Mark 16/22 inventory were released into a disassembly basin, gamma radiation levels at the basin surface would be about one Rem/hour.

** The exception is 432 Mark 22 fuel assemblies that were irradiated briefly in 1992 during K-Reactor startup. This is the K-14 fuel charge, discussed later.

	Mark 16 and 22	Mark 31	Total
Number of Assemblies	586 and 1296	418	-
Initial Heavy Metal Mass (Metric Tons)	6.1	140	146
Fission Product Activity (M Curies)	19.6	0.87	20.5

Table 2:   Summary of SRS Defense-Production Irradiated Fuel and Target Assemblies (Mark 16/22/31) in K-Basin, L-Basin, P-Basin, H-Canyon and F-Canyon. For the Mark 31, one assembly contains 18 slugs. The basis for Table 2 is provided in the Appendix.

B.   Facility Description

As discussed, the facilities currently utilized in the storage of the Mark 16/22 spent fuel include the Reactor Disassembly Basins, the H-Canyon Storage Basin, and the RBOF. These facilities are briefly described below.

Reactor Disassembly Basins: Since 1954 the K-Basin, L-Basin, and P-Basin have been used for disassembly and storage of irradiated fuel assemblies and targets. The basins are functionally identical and similar in construction, although one basin is larger than the other two (P-Basin is 4.6 million gallons while K-Basin and L-Basin are 3.4 million gallons). Each basin's depth ranges from 17 to 50 feet.

The four main sections for fuel disassembly and storage are as follows:

- The vertical tube storage section (VTS): When the reactors were operating, irradiated assemblies were moved from the reactor process room to the VTS. In the VTS, the assemblies were suspended individually from stainless steel hangers and a monorail system until decayed sufficiently for disassembly.

- The machine area: When sufficiently decayed, individual assemblies were moved from VTS to the machine area where the fuel was mechanically separated from targets and bundled.

- The horizontal bundle and bucket storage area (HBBS): After mechanical separation, fuel bundles were moved to the HBBS and stored in horizontal aluminum racks (alloy 6061).

- The transfer area: After a period, fuel bundles were moved to the transfer area (consisting of two water-filled pits) and then loaded into submerged shipping casks, and shipped to the chemical processing area.

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The basins were constructed of reinforced concrete to standards in effect in the 1950s. The basins are below grade, epoxy-lined or painted, and designed to withstand a 1000 psf blast load and a nominal 0.1 g earthquake. In the basin's Basis for Interim Operation,[7] WSRC indicates that the basin's exterior walls and foundations "should be capable" of withstanding a 0.2 g earthquake, based on a scoping study, but no additional analysis exists to qualify the basins.

In the fall of 1993, the DOE Spent Fuel Working Group reported assessments of 66 facilities storing spent fuel and reactor irradiated nuclear material at 11 DOE sites. The report concluded that the L-Basin and K-Basin at the SRS were two of the five facilities with the most significant vulnerabilities.[6] This report also identified vulnerabilities not discussed in the Basis for Interim Operation, including seismic vulnerabilities with interior basin structures and systems that could result in fuel rupture. The study states repeatedly that processing delays or longer term storage would require authorization basis upgrades, seismic evaluations, water chemistry control upgrades, and facility life extension plans. Specific potential vulnerabilities associated with long-term fuel storage in these basins include the following:

- The basin's structure above the base mat is essentially two segments, separated by an expansion joint with a water stop. The base mat is also discontinuous at this joint. Because of mass and stiffness differences, the two segments will respond differently in an earthquake, potentially failing the water stops and causing the basin to leak at an unpredictable rate.

- The vertical frames in the vertical tube storage area lack adequate steel reinforcement to withstand a design basis earthquake. Since these frames support the fuel assemblies, frame failure could lead to dropping the fuel and loss of configuration control (i.e., a criticality concern).

- Since cranes and supports over the basins have not been seismically qualified, these items could fail and fall on fuel in the basin during a design basis earthquake. Also, some other components (e.g., the cask handling crane) may fail and puncture the basin and lead to leakage.

- The ion exchange capacity is not sufficient to maintain high water quality with low activity. WSRC plans to increase ion exchange capacity.

- Leakage through the basin's walls (and the water stops discussed above) can occur even under normal conditions. For example, the L-Basin's concrete wall adjacent to the reactor building has been reported to have cracks that leak.

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- The basins do not have a system capable of detecting small leaks. Although installation of additional monitoring wells outside the basins is planned, these may not be able to detect small leaks due to masking by previous releases.

- The basin's ventilation systems lack exhaust confinement features, such as High-Efficiency Particulate (HEPA) filters.

H-Canyon Storage Basin: At the time of the Spent Fuel Vulnerability Study (November 1993), thirteen Mark 16 fuel assemblies were stored in stainless steel racks in the H-Canyon bundle storage basin. This stainless steel-lined basin (13,000 gallon capacity) has been in operation since the 1950s. The basin is located in a remotely operated, shielded area and has no provisions for water chemistry controls. Ventilation for the basin is the same as for the entire canyon building. All canyon air is pulled through a sand filter, monitored for activity, and exhausted out a stack with a stack monitor.

The H-Canyon design criteria were similar to those for the disassembly basins. The basic canyon structure may be acceptable for a 0.2 g earthquake. Further evaluations are underway[8].

Receiving Basin for Offsite Fuel: RBOF, in operation since 1963, stores fuel in a concrete pool with a stainless steel bottom and painted sides. RBOF has the remains of two Mark 16 type assemblies. RBOF also contains a large inventory of nondefense production spent fuel that is outside the scope of this report.

RBOF was designed to commercial, nonnuclear structural standards, but includes HEPA filtered ventilation and continuous pool water deionization. Potential RBOF vulnerabilities include nonseismically qualified masonry walls near some pools, a roof susceptible to tornadic missiles, nonseismically qualified fuel storage racks, inadequate pool leak detection, and lack of an up-to-date authorization basis.[6]

C.   Fuel Condition

Before 1989, the typical residence time of irradiated fuel in the disassembly basins was less than 18 months. However, nearly all the irradiated material currently in the basins has now been there at least seven years. The aluminum cladding on fuel is typically 0.030 inch thick. The thinnest cladding possible was used in these designs to facilitate dissolution and subsequent processing. The cladding design was not intended for long-term storage in water. During the extended storage, pitting corrosion of the aluminum clad has occurred. In many cases, the pitting has penetrated the clad into the fuel core and fission products are being released into the basin's water.[9]

Vertical Storage: About one-third of the Mark 16/22 fuel assemblies are in vertical storage in the disassembly basins suspended from stainless steel hangers. Visual inspection of this fuel revealed pitting corrosion of the aluminum cladding with evidence of fuel core corrosion. Pitting has occurred along the entire length of the fuel hanging in vertical storage. Pitting along the length of the fuel is attributed to the low quality water (conductivity of 100-180 μS/cm), and has occurred mainly along scratches that broke through the protective oxide film. Some of the pits have completely penetrated the clad, and corrosion of the fuel core is evident. In addition, an increased amount of pitting has occurred near the top of the fuel adjacent to the stainless steel hanger. The increased pitting near the top of the fuel is attributed to the galvanic contribution from the stainless steel hanger. It should be noted that all of the Mark 16/22 fuel were kept in vertical storage for some time after being discharged from the reactor, and all of the fuel have been exposed to the galvanic influence of the stainless steel hangers.

Horizontal Storage: The Mark 16/22 fuel assemblies in horizontal storage are supported by aluminum racks and do not exhibit the pitting observed along scratches on the vertically stored fuel. This fuel is stored in aluminum bundles inside larger aluminum storage racks. Significant corrosion is visible on the aluminum racks, indicating that the racks, made from a different aluminum alloy than the fuel cladding, may be anodic to the aluminum fuel and provide cathodic protection.

WSRC is installing new horizontal storage racks for the vertically hanging fuel. These racks may provide cathodic protection to the fuel and slow initiation of new pitting on the clad, but will not stop growth of pits already established in the clad or stop corrosion in the exposed fuel core. The cathodic protection from the racks currently in service was not expected or planned. Other factors such as surface finish, surface contamination, or residual stresses may have provided the difference in electrochemical potential. Hence, it is not certain that similar aluminum racks made today will provide the same cathodic protection as the old racks.

Water Quality: One of the key features affecting aluminum pitting corrosion during wet storage is the quality of the water. Most of the commercial spent fuel storage basins in the United States contain high-quality deionized water. High-quality water is achieved by continuous deionization of the water to remove corrosion causing impurities, such as chloride ions, and to maintain water conductivity below 10 μS/cm. At the SRS, only the RBOF was designed for continuous deionization to maintain high-quality water with conductivity less than 10 μS/cm. Intact, undamaged aluminum-clad fuel has been stored for 11 years and aluminum racks for over 30 years in the RBOF without visible signs of corrosion.

The P-Basin, K-Basin, and L-Basin do not have continuous deionization systems and the water quality is poor, with conductivities in the range of 100-180 μS/cm. This high conductivity is caused by a high level of dissolved ions (about 30 ppm) and indicates the water can readily support electrochemical corrosion. Corrosion has not generally been a problem in the past because the fuel was removed within 18 months. However, past excursions in water quality have caused corrosion problems even during these short storage periods.

During the period 1974-1977, there were four occurrences of clad-penetrating corrosion in the K-Basin. In these occurrences, tritium and fission product releases were detected one to three months after the spent fuel was put into the basin.[9] Subsequent inspection revealed extensive corrosion and penetration of the aluminum clad. A number of factors were believed responsible for the corrosion: poor cladding material, excessive iron and chloride ions in the water, galvanic couples, and scratches in the oxide coating. The chloride ion concentration was reduced from 25 ppm to 5 ppm, and the pH was raised slightly to 7.3 to reduce the solubility of heavy metal ions in the water.

After the changes were implemented in 1978, there was no reported corrosion of fuel in the basins until 1990. At this time pitting corrosion of aluminum sleeve housings stored in the K-Basin was observed. This corrosion occurred after only 25 to 70 days in the basin and several pits penetrated the 0.050-inch wall.[10] Aluminum samples and coupons exposed in the K-Basin and L-Basin during 1990 and 1991 confirmed that this pitting was due to poor water quality. The samples showed severe pitting with rates as high as 0.002 inch/day. A pitting rate of 0.002 inch/day would penetrate the typical fuel cladding in 15 days. Testing similar samples in the high quality water of RBOF revealed no pitting. Water conductivities during these tests were 1 μS/cm in RBOF versus 178 μS/cm in K-Basin.

In an effort to improve water quality, WSRC plans to clean the basins and install continuous filtering.[11] A concerted effort was initiated in 1993 to clean up P-Basin and K-Basin. Deionization of the water began by using portable mixed-bed deionizers, and the conductivity and impurity concentration of the water has been gradually lowered. Subsequent corrosion testing during 1993-1995 confirmed the absence of pitting on aluminum samples in K-Basin and L-Basin. The remarkable improvement in pitting resistance was attributed by WSRC to a slight reduction in conductivity of the water (178 μS/cm reduced to 125 μS/cm). Test samples exposed in the P-Basin during this same period showed no pitting through eight months. After 11 months, however, pits as deep as 0.035 inch were found. At the time of pitting, the P-Basin had not been deionized for over two years, and the conductivity was 165 μS/cm.

Another mechanism of corrosion that has not been specifically identified in SRS basins is microbiologically influenced corrosion (MIC). MIC of aluminum is associated with bacteria or other microorganisms. The bacteria do not cause the corrosion but create the localized environment that enables electrochemical corrosion to be accelerated. Unlike other corrosion processes, MIC is not often visible because it does not usually generate extensive corrosion products.

MIC was identified as the cause of pitting corrosion on aluminum coupon specimens and aluminum cans in the CPP-666 Basin at the Idaho National Engineering Laboratory.[9] The water quality in the CPP-666 Basin is among the best of all the DOE nuclear fuel storage basins. With a low chloride concentration and a low conductivity, corrosion of aluminum-clad fuels has not been a problem. However, during an inspection in 1992, pitting corrosion was observed on aluminum coupons in the basin. These coupons were found to have a biofilm covering the pits. In 1993 a dummy aluminum fuel storage canister was found to have pits that resulted in penetration of the 1/8-inch thick wall. The pits contained no corrosion products and could not be seen with the naked eye a few feet away. The corrosion was attributed to MIC. MIC has the potential to play a role in the corrosion of spent fuel during continued storage at the SRS.

The P-Basin, K-Basin, and L-Basin are painted concrete basins that may preclude achieving the high quality water found in the newer stainless steel lined basins. However, any improvement in water quality should slow, and may even prevent, the initiation of new pitting in the clad. In the 94-1 Implementation Plan, DOE reported that corrosion coupons tested in the basins between 1990 and 1994 indicated progress had been made in slowing the corrosion rates of aluminum in the basins. However, this does not mean that the aluminum corrosion of SRS spent fuel has been significantly reduced or stopped. In fact, existing pits are not expected to be strongly influenced by water quality improvements and are expected to continue corroding. As a result, corrosion and release of radioactive contaminants into the water are expected to continue during wet storage regardless of water quality.

Moreover, the variations in conductivity between 100-180 μS/cm cannot solely explain the outbreaks of extremely aggressive pitting observed in these basins. WSRC scientists have reported that factors promoting the corrosion of aluminum are complex, not well understood and, in many cases, operate synergistically.[9] Among the factors believed to promote pitting corrosion are:

- High water conductivity.

- Aggressive ions (such as chlorides and sulfates).

- Sludge (containing high concentrations of iron and chlorides).

- Galvanic couples.

- Scratches and imperfections in the oxide coating on the clad.

- Relatively stagnant water.

- Microbiologically influenced corrosion (MIC).

As a result of these complexities, it is oversimplistic to assume that improvements in water quality will guarantee future fuel integrity. In addition, excursions in water quality similar to those that occurred in previous years could occur again. In either case, more aggressive pitting corrosion and large releases of radioactive contaminants are possible with continued wet storage.

IV. REMEDIATION ALTERNATIVES

A. Chemical Processing

Reactor-irradiated nuclear fuel has been processed at the SRS for over 40 years using proven, existing technologies and facilities. H-Canyon began operation in 1955 and was originally designed to recover plutonium and uranium from reactor-irradiated natural uranium fuel using the Plutonium Uranium Extraction (PUREX) process.[8,12] Several years later, the H-Canyon process was modified (the HM process) to permit recovery of neptunium and enriched uranium. The process flowsheet was also modified to reduce criticality risk.

The use of the H-Canyon to process the Mark 16/22 fuel to low-enriched oxide is depicted in Figure 1 and described below. A variation of this process, using the F-Canyon to process the fuel, is also described.

H-Canyon Processing: The H-Canyon building contains two parallel canyons designed to isolate the processing equipment from operating personnel. These canyons allow for remote, highly radioactive operations.
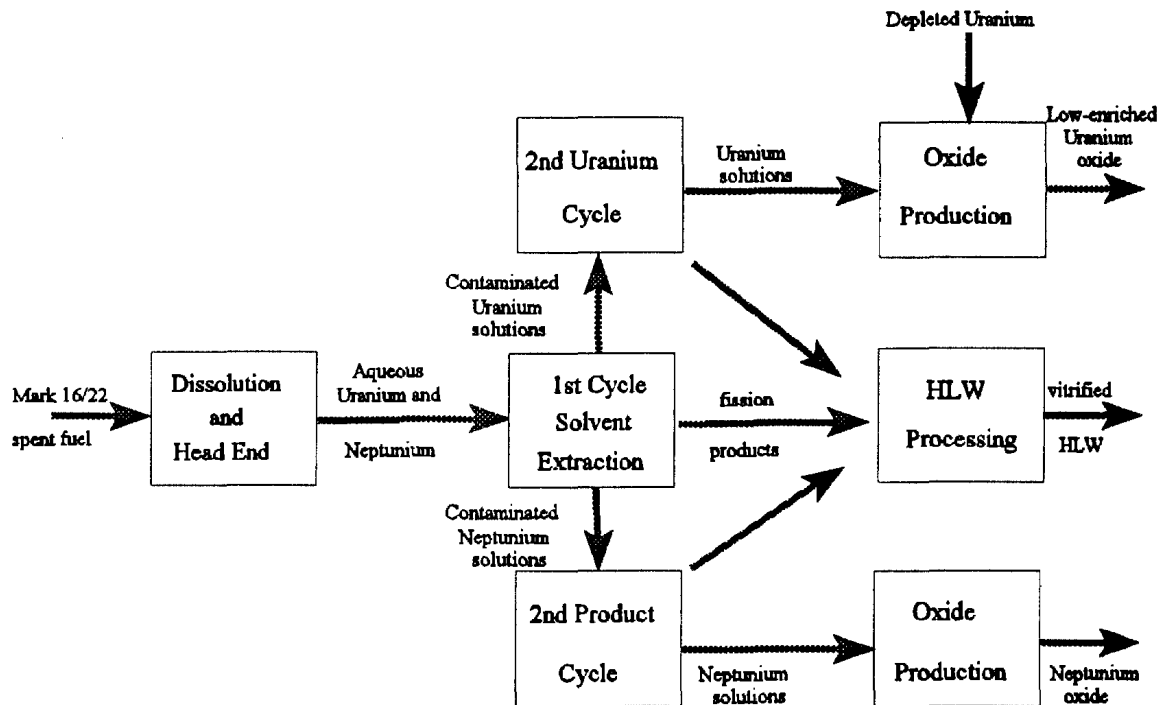

Figure 1:   H-Canyon Fuel Processing.

The first processing step in H-Canyon is dissolution of the fuel. The fuel is dissolved either chemically in a boiling bath of nitric acid or anodically in an electrolytic dissolver. Following dissolution, the aqueous solution is clarified to remove precipitated impurities and fission products in the head end section of the process. The cleaned solution is then transferred to the solvent extraction section. The actinides (uranium, neptunium, and any trace amount of plutonium) are extracted in the organic solution while the fission products and aluminum remain in the aqueous stream. This aqueous stream is concentrated and transferred to the high-level waste tanks. The organic solution proceeds to the uranium/neptunium separation bank.

The actinide organic stream is contacted with dilute nitric acid containing a reductant. The neptunium and any trace plutonium are stripped into the aqueous stream and are sent to the second product cycle for further processing. The uranium is back-extracted in dilute nitric acid to an aqueous phase and is purified in the second uranium cycle. For processing of the Mark 16/22 fuel, the purified uranium solution will be isotopically diluted with depleted uranium and converted to a low-enriched oxide. The neptunium solution, following purification in the second product cycle, will be converted to neptunium oxide.

The high-level waste generated as a result of chemically processing the Mark 16/22 fuel consists primarily of aqueous solutions contaminated with fission products and trace quantities of fissile material. Additionally, spent organic solution and some insoluble solids contaminated with high-level waste will require disposal. The high-level waste from Mark 16/22 chemical processing is similar to high-level waste produced during historical operations at the SRS. Processing high-level waste at the SRS involves the Waste Tanks, the Defense Waste Processing Facility (DWPF), and the Saltstone Facility. Using an oxide loading of 27 weight percent per DWPF canister, the high-level waste volume resulting from chemical processing of Mark 16/22 fuel has been recently estimated at 32 canisters.[11] This estimate is slightly more conservative than the IMNM EIS prediction of 22 DWPF canisters. The estimated number of canisters resulting from processing the Mark 16/22 fuels equates to less than one percent of the predicted total number of canisters generated from processing historical SRS high-level waste.

F-Canyon Processing: With some minor processing flowsheet modifications, irradiated fuel can also be processed in F-Canyon. F-Canyon utilizes the PUREX solvent extraction process to separate uranium and plutonium from irradiated depleted uranium targets (Mark 31s). The two canyon structures are alike and the F-Canyon PUREX process is very similar to the H-Canyon HM process. One difference between the two processes is in the second product cycle where the PUREX process recovers plutonium while the HM process recovers neptunium.

The primary changes required in F-Canyon for fuel processing involve criticality control and the second product cycle. For criticality control, inserts will be placed in the dissolvers and the dissolved solutions will be isotopically diluted with depleted uranium prior to solvent extraction. The depleted uranium may be supplied from depleted uranium solutions or oxides in inventory. The second product cycle will require flowsheet modifications to extract neptunium instead of plutonium. The trace plutonium in the dissolved fuel will be sent to high-level waste processing along with the fission products.

The K-14 low burnup Mark 22 core will require only limited processing in F-Canyon. The K-14 charge, because of the short irradiation time and long storage, has a very low fission product activity and negligible neptunium concentration when compared to the higher burnup cores. For these reasons, processing the K-14 core in F-Canyon may not require the second uranium cycle or neptunium recovery.

B.   Dry Storage

Although the experience with dry storage of aluminum-clad spent nuclear fuel is very limited, there is general consensus that undamaged spent fuel of this type can be stored safely in a dry condition. However, in order for the damaged Mark 16/22 fuel to be placed safely into 30-50 year dry storage, technical and regulatory acceptance criteria must be developed, new facilities must be designed and constructed, and packaging and stabilization activities must be performed. In addition, criteria for ultimate disposition, such as waste acceptance criteria for disposal in a geologic repository, need to be addressed to minimize the potential for having to handle, package, or process the fuel in the future.

Previous Experience: Unlike commercial type zircaloy-clad uranium-oxide fuel, the experience with dry storage of aluminum-alloy fuel is very limited. In fact, while at least 13 countries have experience in the dry storage of commercial type nuclear fuel, only four instances of dry storage of aluminum fuel are generally identified, and no experience with dry storing previously damaged aluminum-alloy fuel could be found. This limited experience with dry storing undamaged aluminum fuel is summarized below.[13]

- The Lucas Heights Facility in Australia has stored aluminum-alloy spent nuclear fuel in sealed, stainless steel tubes that are inserted into 50 foot deep boreholes in the ground. The atmosphere in these sealed, stainless steel tubes is dry nitrogen. Fuel examined from this storage facility after 10 and 25 years of storage showed no visible signs of corrosion.

- At the Japan Atomic Energy Research Institute, aluminum-clad uranium metal and uranium-oxide fuel are stored in sealed, stainless steel canisters that are inserted into stainless steel lined drywells imbedded in a large concrete block. The atmosphere in the sealed, stainless steel canister is helium. The drywell atmosphere is maintained at a subatmospheric pressure, and is kept dry using dehumidifiers. Fuel at this facility, examined after five years of storage, showed no signs of degradation.

- At Los Alamos, 48 Materials Test Reactor aluminum-alloy fuel elements are being dry stored in a transportation cask. This fuel has been stored in these casks for about three years. These fuel elements, visually inspected after two years of storage, showed no visible signs of corrosion.

- At the Chalk River Laboratory in Canada, some pin-type aluminum clad fuel has been stored in vented steel-lined drywells. These vertical drywells are about 16 feet high and are buried in sand. The condition of the fuel in these drywells is not known.

No degradation of the fuel clad prior to storage is noted for these specific cases, and it is reasonable to assume that this is the case. This very limited dry storage experience seems to indicate that undamaged aluminum fuel can be successfully dry stored. Also, the very limited inspection results of dry stored aluminum spent fuel seem to be consistent with industrial reports of aluminum alloy corrosion in the successful formation of a passive oxide layer. These reports result from 30-year tests of aluminum alloys exposed to industrial seacoast environments.[14] Examination of these alloys revealed maximum pitting depths of about 0.010 inches, or about one-third the thickness of the typical cladding of Mark 16/22 fuel elements. These results in industrial environments, however, have not been validated for irradiated materials, and do not include the potential adverse effects of previous basin water exposure on the protective oxide films.

Technical and Regulatory Acceptance Criteria: There are significant differences between the aluminum-clad, aluminum-alloy fuels at the SRS and the zircaloy-clad UO_2 ceramic commercial spent fuel being held in dry storage at several locations across the country. The more obvious differences include the lower melting temperature of aluminum, the more corrosive nature of aluminum, the substantially higher enrichment of the Mark 16/22 aluminum fuel, the lower heat generation rates of the aluminum fuel, and the degraded condition of the aluminum fuel. Some of these differences present uncertainties that have to be addressed in developing storage criteria. For instance, in performing the criticality assessment for dry aluminum fuel, the lack of experimental data raises questions of the validity of applying current cross section libraries to a dry, fast system of aluminum and uranium.[15]

The 10 CFR 72 general design requirement addressing confinement barriers and systems states that "[t]he spent fuel cladding must be protected during storage against degradation that leads to gross ruptures, or the fuel must be otherwise confined such that degradation of the fuel during storage will not pose operational safety problems with respect to its removal from storage." Compliance with this requirement demands an understanding of the potential degradation mechanisms and development of technical and regulatory acceptance criteria.

These acceptance criteria would have to address maximum fuel temperature and storage environment requirements, such as maximum humidity and requirements for inert gas. Defining these acceptance criteria requires an understanding of the temperature effects and material compatibility on fuel degradation, the heat generation and heat removal properties, and the consequences of existing corrosion or other cladding damage.

In addition, some amount of fuel characterization is required for dry storage. It is likely that additional characterization will be needed to support subsequent long-term or permanent disposal. Future handling of the fuel may be required for additional characterization if the fuel is not adequately characterized prior to placement in interim dry storage.

The interim dry storage alternative could only be practical if it was followed directly by long-term or permanent disposal. One of the potential problems with ensuring this is the issue of criticality safety in a geologic repository. For commercial spent fuel, recent emphasis has been placed on containing fuel in a multipurpose canister (MPC) with poisoned dividers and jacketed by an overpack for final disposal in a repository. Initial assessments conclude that for aluminum-clad highly enriched uranium fuel, the success of the MPC approach cannot be guaranteed because the fuel is subject to disintegration in dry storage at a rate determined by previous history and water quality.[17] Thus, the material problems and uncertainties with the aluminum fuel affect the probabilities and risk of criticality events in the repository. Current programs to address these uncertainties include development of alternate waste forms that would avoid these uncertainties if the risks associated with the MPC-type waste form be judged unacceptable. Unfortunately, some of the possible waste forms being considered require extensive processing. Several packaging strategies for preventing or delaying criticality are briefly described below:[16]

- Mass Limitation: This strategy involves the simple process of limiting the amount of fissile material in each package. The possibility that the fissile material might be dissolved, transported, and collected elsewhere to form a critical mass is reduced, but not eliminated. The most significant impact of this strategy is the potential to increase the disposal costs by increasing the number of waste packages produced.

- Neutron Poison: This strategy requires the fuel to be packaged at a relatively high density through vitrification, melting, mechanically chopping, crushing, shredding, or by simply close packing the fuel. Criticality would then be controlled by adding a neutron poison such as gadolinium.

- Isotopic Dilution: This is also a strategy for packaging the fuel at a relatively high density. Again, the fuel would be packaged through vitrification, melting, mechanically chopping, crushing, shredding, or by simply close packing the fuel. However, instead of introducing a neutron poison into the package, criticality would be controlled by isotopic dilution obtained from adding depleted uranium.

Figure 2 has been taken from the DOE Spent Nuclear Fuel Technology Integration Plan.[17] This figure shows the actions resulting from both the dry storage alternative and the chemical separation alternative. The chemical processing alternative, denoted as "Condition (Process, Direct Dispose, etc.)" in Figure 2, faces only one criteria-driven qualification step to reach ultimate disposal. DOE has expended considerable effort qualifying the vitrified high-level waste product to be produced at the SRS. On the other hand, the dry storage route shown in Figure 2 involves at least five criteria-driven actions and will require significant development work. Figure 2 also illustrates the possibility, given that the spent fuel is not acceptable for disposal in a geologic repository, for subsequent processing of the fuel.

Processing and Storage Facilities: The current concept for dry storage of the Mark 16/22 fuel at the SRS would involve storage in either a modular dry storage vault or in multiple dry storage casks. Neither of these storage capabilities currently exist at the SRS and would have to be designed, constructed, and put into operation.

In addition to various support components that may be required by the design (such as a wet pool for fuel receipt, and remote or shielded handling apparatus), a modular dry storage vault would consist primarily of a processing component and a vault storage component. Previous designs for the vault storage component utilize a massive underground concrete vault (5 feet thick walls) where vertical steel tubes are installed to hold canisters of spent fuel. The massive concrete vault provides the required shielding and most of the structural resistance to natural hazards such as earthquakes. The vertical steel tubes provide some of the configuration control of the spent fuel to prevent potential criticality and to allow space for heat removal. Confinement of the spent fuel is provided by the fuel assembly, the fuel canister, the vertical steel storage tube, and the concrete vault and associated ventilation system.
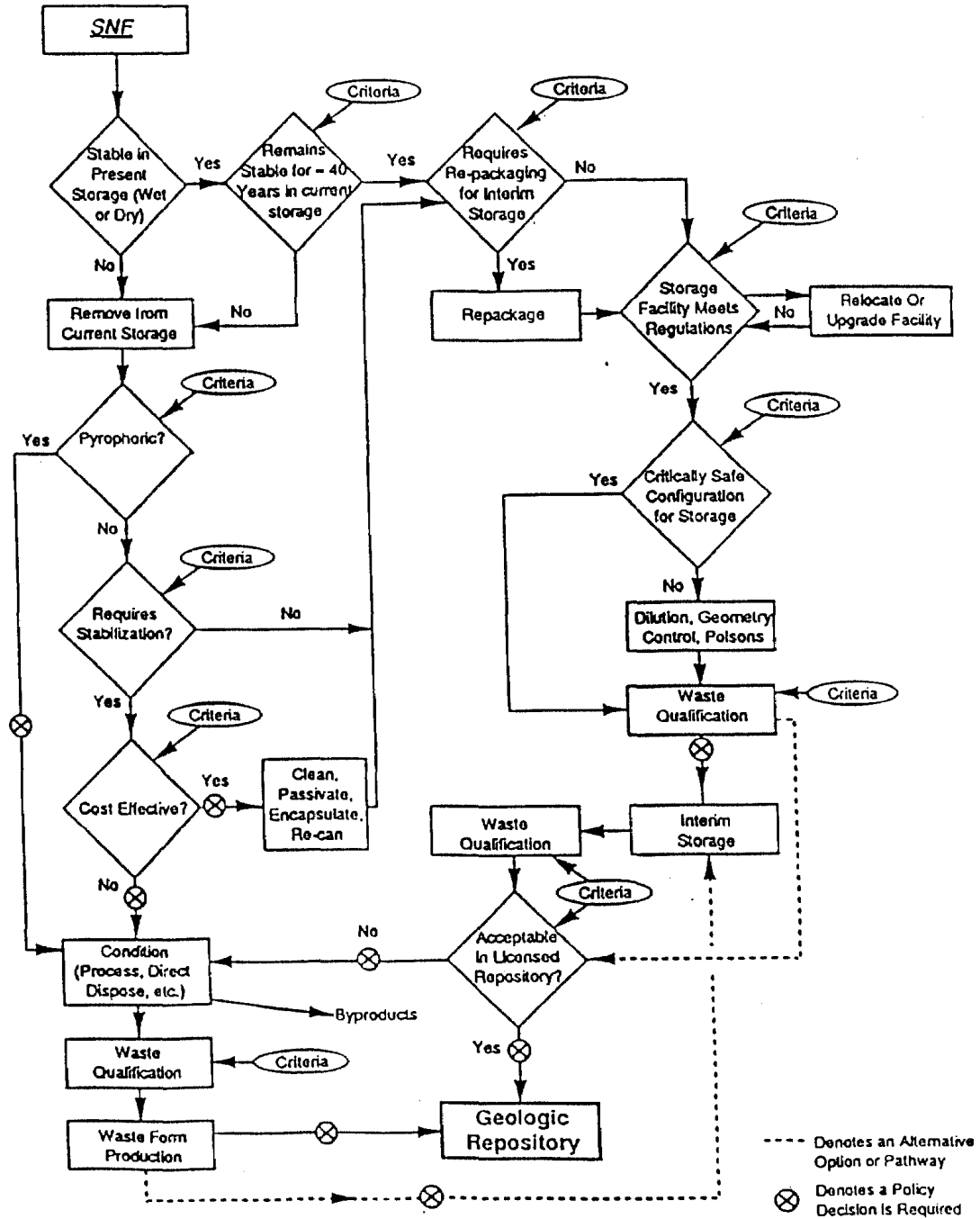
Figure 2: Flow Diagram of Activities Needed to Achieve Ultimate Disposal of DOE-Owned Spent Nuclear Fuel in a Geologic Repository.[17]

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Several steps are required to place fuel into the passive dry storage condition. The first step requires handling the fuel in the current wet storage basins, and performing some initial configuring and packaging of the fuel into the new dry storage canisters. Currently, DOE has determined that, precluding facility modification, only the RBOF facility has the capability for loading and inspecting fuel under water. With some modifications, it is reported that L-Basin could obtain this capability. In any case, the fuel would be placed in a fuel storage canister, and a neutron-absorbing material might be added for criticality control.

The most prevalent conceptual designs for dry vault storage of spent fuel involve sealed fuel storage canisters. The sealed canister provides the first reliable barrier between the spent fuel and the environment (particularly for fuel with known cladding failure or degradation). Prior to sealing a canister containing spent fuel, issues such as continued corrosion, gas generation, radiolysis, etc., have to be addressed. These issues are resolved by the drying and stabilizing of the fuel and the inerting of the canister. In this process, water is drained or purged from the canister, and the fueled canister is processed to remove the remaining moisture and any other reactive attributes.

The irradiated Mark 16/22 fuel is considered high-level waste due to the fission product content. Dry storage will not provide high-level waste volume reduction because the fission products remain entrained in the fuel elements. SRS personnel estimate that 46 dry storage casks will be required to contain the Mark 16/22 fuel. Using the SRS factor of four DWPF canisters per dry storage cask, the high-level waste volume associated with dry storage is equivalent to 184 DWPF canisters.

Other Requirements: In describing the dry storage alternative in the draft IMNM EIS, DOE indicates that this alternative would require the construction of a new facility on a previously undisturbed site at the SRS. Construction and operation of this new dry storage facility would be evaluated through the preparation of a project-specific environmental assessment or, more likely, an EIS. In addition, the dry storage alternative creates a new operation at the SRS consisting of maintenance, surveillance, and inspection of the new high-level waste form and its processing and storage facility.

V. COMPARISON OF ALTERNATIVES / CONCLUSIONS

In comparing the merits of chemically processing the fuel to a low-enriched oxide and placing the fuel in dry storage, several conclusions can be reached. These conclusions are elaborated below.

A. Stabilization of the fuel by chemical processing to a low-enriched oxide reduces the risk associated with continued wet storage and subsequent dry storage of the deteriorating fuel. As long as the fuel remains in wet storage, the risk will continue to increase at an uncertain rate due to corrosion. The degraded spent fuel will continue to be susceptible, not only to corrosion at existing cladding breaks, but also to short-term water chemistry excursions and other unanticipated effects that could accelerate clad failure and fission product release. The risks of continued wet storage include:

- Increased vulnerability due to potential leakage and seismic vulnerabilities.

- Increased radiation exposure to workers if increased basin water fission product activity results in radioactive deposits at the waterline (similar to the Hanford K-East basin's "bathtub ring"). Worker radiation exposure (as well as radioactive waste volume) may also increase if water filter media changeout is required more frequently to maintain low water activity levels.

- Increased radioactivity in basin sludge that makes water quality control difficult and constitutes a risk of environmental insult in the event of basin leakage.

Stabilization of the fuel by chemical processing will remove the fuel from wet storage by 1999. The dry storage option does not remove the fuel from the basins until 2003-2008, thus increasing the risks associated with wet storage for another 4 to 9 years. In addition, the dry storage option has much greater technical uncertainty that might further extend the time in wet storage.

The risk associated with chemical processing is well known and has been quantified.[8,12,21,22,23] If the canyon processing option is chosen, there is initially a higher relative risk, but this risk is reduced in a short time (i.e., three years).

B. Stabilization of the fuel by chemical processing to a low-enriched oxide eliminates the need for additional processing or handling activities in the future that would involve additional safety risks. After an interim period of dry storage (30 to 50 years), the aluminum-clad fuel is to be transferred to ultimate disposal under the commercial spent nuclear fuel program. There has not been significant development of waste acceptance criteria for highly enriched aluminum-clad fuel in a geologic repository (particularly for damaged fuel). Questions of fuel disintegration and criticality have not been explored. Extended wet storage and additional degradation of the fuel increase uncertainty in the development of waste acceptance criteria for the Mark 16/22 fuel. The condition of the fuel and the activities that would be required in the future are unknown. The possibility exists for having to perform future processing, repackaging, or some other treatment to allow disposal. Safety risks associated with these potential future activities are likely to equal or exceed those risks associated with near-term chemical processing.

C. Stabilization of the fuel by chemical processing to a low-enriched oxide produces the smallest volume of high-level waste. The estimated high-level waste from dry storage of the Mark 16/22 fuel is equivalent to 184 DWPF canisters. By comparison, chemical processing of the Mark 16/22 fuel is estimated to produce 32 high-level waste canisters. This represents an 82 percent volume reduction of high-level waste when compared to the dry storage option, and is less than one percent of the estimated total number of DWPF canisters to be produced from historical SRS high-level tank waste.

D. Stabilization of the fuel by chemical processing to a low-enriched oxide is the lowest cost alternative. Cost information is not provided in this report. However, several assessments have been performed to compare costs between the dry storage and chemical processing alternatives for stabilizing SRS aluminum spent fuel.[11, 18]

APPENDIX - SRS Spent Fuel Inventory

The SRS Team Report in the Spent Fuel Vulnerability Study[6] provides an inventory of Mark 16/22 spent fuel assemblies and Mark 31 target assemblies (18 slugs per assembly). This inventory is summarized in Table A1. The SRS Disassembly Basin Hazard Assessment Documents[19, 20] provide additional information on the heavy metal mass and the fission product activity at reactor shutdown for the inventories in L-Basin and P-Basin. Using that data, the heavy metal and fission product inventories (decayed to seven years after shutdown) in Tables A2 and A3 were developed.

The Board's staff is aware of two Mark 16 assemblies in the Receiving Basin for Off-Site Fuel (RBOF) that are reflected in the following tables. RBOF has a large inventory of nondefense production irradiated material that is not included in the tables below.

Facility	Facility Area	Mark 22 Assemblies	Mark 16 Assemblies	Mark 31 Assemblies
K-Basin	Vertical Tube Storage	432	-	-
	Horizontal Tube Storage	468	1	-
	Bucket Storage	-	-	3
L-Basin	Vertical Tube Storage	-	252	-
	Horizontal Tube Storage	-	265	-
	Bucket Storage/Machine Area	-	-	347
P-Basin	Vertical Tube Storage	-	-	-
	Horizontal Tube Storage	396	53	-
	Bucket Storage	-	-	-
RBOF		-	2	-
H-Canyon	hot canyon basin	-	13	-
F-Canyon	hot canyon basin	-	-	68
Total		1296	586	418

Table A1:   Inventory of Defense-Production Irradiated Fuel and Targets (Mark 16/22/31).

Facility	Mark 22	Mark 16	Mark 31	Total
K-Basin	2.89	0.003	1.00	3.89
L-Basin	-	1.72	116.2	117.9
P-Basin	1.27	0.18	-	1.45
RBOF	-	0.003	-	0.003
H-Canyon	-	0.04	-	0.04
F-Canyon	-	-	22.8	22.8
Total	4.16	1.94	140	146

Table A2:   Initial Heavy Metal Mass (Metric Tons) in SRS Defense-Production Irradiated Fuel and Targets.

Facility	Mark 22	Mark 16	Mark 31	Total
K-Basin	8.32	0.01	0.01	8.34
L-Basin	-	6.72	0.72	7.44
P-Basin	3.66	0.69	-	4.35
RBOF	-	0.01	-	0.01
H-Canyon	-	0.17	-	0.17
F-Canyon	-	-	0.14	0.14
Total	11.98	7.60	0.87	20.45

Table A3:   Fission Product Activity (M Curies) in SRS Defense-Production Irradiated Fuel and Targets.

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