

WSRC-TR-96-0401 (U)

# CRITERIA FOR ACCEPTANCE OF OFF-SITE SPENT NUCLEAR FUEL AT SRS (U)

R. L. Sindelar and J. P. Howell

Savannah River Technology Center  
Strategic Materials Technology Department  
Materials Technology Section

Publication Date: December 1996

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This document was prepared in connection with work done under Contract No. DE-AC09-96SR18500 with the U. S. Department of Energy

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**DOCUMENT:** WSRC-TR-96-0401  
**TITLE:** CRITERIA FOR ACCEPTANCE OF OFF-SITE SPENT NUCLEAR FUEL AT SRS (U)  
**TASK:** FOREIGN FUEL RECEIPT CRITERIA IMPROVEMENT PROGRAM, REF. EFR-960052

**APPROVALS**

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*R. L. Sindelar* Date: 12/18/96  
R. L. Sindelar, Author  
Materials Applications & Corrosion Technology Group  
MATERIALS TECHNOLOGY SECTION

*J. P. Howell* Date: 12/18/96  
J. P. Howell, Author  
MATERIALS TECHNOLOGY SECTION

*M. C. Thompson* Date: 12/18/96  
M. C. Thompson, Technical Reviewer  
CHEMICAL & HYDROGEN TECHNOLOGY SECTION

*N. C. Iyer* Date: 12/20/96  
N. C. Iyer, Manager  
Materials Applications & Corrosion Technology Group  
MATERIALS TECHNOLOGY SECTION

*T. L. Capeletti* Date: 12/20/96  
T. L. Capeletti, Manager  
MATERIALS TECHNOLOGY SECTION

*S. D. Burke* Date: 12/18/96  
S. D. Burke, Manager  
Basin Management  
SPENT FUEL STORAGE DIVISION

---

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## 1.0 SUMMARY

Criteria have been developed for the acceptable condition of aluminum spent nuclear fuel (SNF) for direct storage in water basins at the Savannah River Site (SRS). These criteria are recommended for future receipts of aluminum SNF from foreign research reactors (FRR).

### 1.1 Visual Examination and Acceptance Criteria

Visual examination of the fuel elements of each fuel assembly shall be performed at the FRR storage basin prior to loading for shipment to SRS. The regions of the fuel assembly to be examined are those outside surfaces of the fuel elements directly observable through basin water. The acceptance criteria for the examination are:

- 1) No exposure of the fuel meat as a result of i) mechanical damage or cutting, ii) through-clad cracks, or iii) localized corrosion or pitting; and
- 2) No corrosion nodules on the cladding over the fuel meat region greater than approximately 1/8" in diameter.

Fuels whose condition meets the criteria would have a low potential of having a through-clad penetration which could result in release of fuel or fission product species from the fuel meat during shipping to and storage in the Savannah River Site (SRS) basins. These fuels are acceptable for direct storage in SRS basins. Fuels whose condition does not meet the criteria would have to be encapsulated prior to storage in SRS basins.

### 1.2 Radionuclide Sampling Criteria

Radionuclide release rates can be determined by measuring radioactivity in the water while the fuel assembly "rests" in a container of water for a period of time. This radionuclide sampling technique is commonly referred to as a sip test [1]. Experience has shown that visual examination is more sensitive in detecting potential cladding breaches in the aluminum-based research reactor fuel [2]. Furthermore, the results of sip tests may be strongly influenced by surface contamination. Performing a sip test with the existing interim criteria [3, 4] is recommended as an option in order to build a technical database needed to support future decisions on encapsulation, shipping, and storage of aluminum-clad spent nuclear fuel.<sup>1</sup>

## 2.0 INTRODUCTION

The SRS will be receiving aluminum SNF from FRR to implement the U. S. Department of Energy (DOE) Record of Decision for the FRR Environmental Impact Statement [5]. The SNF is to be temporarily stored for up to 15 years in the Receiving Basin for Offsite Fuels (RBOF) and the L disassembly basin.

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<sup>1</sup>A maximum release rate of 13  $\mu\text{Ci/hr}$  of Cs-137 per fuel shipment was recommended for fuel acceptance [3,4] for direct basin storage to assure that administrative limits for activity in the basin water are maintained under existing basin deionization capacities.

This report provides a criteria and the necessary technical basis for accepting future receipts of aluminum SNF from FRR for storage in the SRS basins without encapsulation. The criteria is based on the use of a visual examination to identify SNF that have a high potential for having breached cladding. Guidance for implementation of the examination requirements in the field is provided.

The release of radioisotopes of SNF from aluminum-based fuel into basin water is reviewed and the performance of fuels with postulated breached cladding in direct storage in SRS basins is evaluated.

Additional recommendations for extended basin storage of fuels not meeting the visual examination criteria are provided. Revisions to this acceptance criteria may be issued as the sip test database and visual examination results become available.

### **3.0 DEVELOPMENT OF CRITERIA FOR DIRECT BASIN STORAGE**

The primary purpose for the cladding on reactor fuel is to seal the fuel meat so that the fuel and radioactive byproducts of nuclear fission are retained in a controlled location. Breaches of the cladding and releases can occur during reactor operation or during post-irradiation handling, transportation, or storage.

The release rates from breached research reactor fuel is much higher during reactor operation than in the storage basins at SRS due to the difference in fuel temperatures (fuel meat temperatures up to approximately 200°C vs. down to room temperatures, respectively) and the kinetics of release. The radionuclide release rate into water at basin conditions, obtained from radionuclide sampling, could be used as a performance indicator to determine acceptability for direct storage of SNF, including those with cladding breaches. The radionuclide sampling test is commonly referred to as a sip test [1].

Evaluating the condition of the cladding can also be performed through visual examination. This method cannot be used to measure radioactivity release, but is more sensitive than the sip test to identify aluminum SNF that have a high potential for cladding breaches.

The following sections are a review of the experience in detecting breaches in aluminum SNF. The requirements for visual examination and the acceptance criteria for storage of FRR SNF in SRS basins without encapsulation are developed. The objective of the visual examination is to identify breaches or potential breaches in the cladding of the SNF. The premise in the development of the acceptance criteria is to not allow storage of FRR SNF with cladding breaches in SRS basins without encapsulation.

#### **3.1 Condition of Aluminum SNF in Basin Storage**

Most of the SNF assemblies from research reactors are being stored in water-filled basins in the U. S. and around the world. The fuel assemblies are predominantly the Materials Test Reactor (MTR) design with aluminum cladding and uranium-aluminum alloy fuel. The thickness of the cladding is nominally 0.015 inches and the fuel core consists of UAl<sub>x</sub> particles dispersed in an aluminum matrix. Some of this fuel has been in water storage for longer than 35 years and the water quality, in several cases, has been less than optimum during some portion of this storage time. This extended storage and less than optimum water quality has resulted in pitting corrosion of some of the fuel assemblies. The corrosion process results in nodular white corrosion products on the cladding surfaces of

the fuel elements of the assemblies which can be observed by visual inspection in the basins.

### 3.2 Pitting Corrosion of Aluminum-Clad Fuel

Irradiated aluminum clad fuel has an aluminum oxide coating ranging from a few, up to approximately 50  $\mu\text{m}$ , in thickness depending on the conditions of irradiation. The aluminum-clad fuel plates may appear grayish or off-white in color which is due to the presence of a film which is generally protective against corrosion [6]. Fuel handling activities can cause scratches in the coating and provide sites for initiation of pitting corrosion. Laboratory corrosion testing has established the role of water composition and conductivity on pitting initiation of aluminum alloys [7]. Many of the studies by Aziz, et al have used laboratory corrosion testing combined with extreme value statistics to derive empirical relations between water composition, conductivity, etc. and the maximum depth of pitting [8]. Some models are better than others, but even these do not accurately predict pitting in deionized, low conductivity water which is typically used in most fuel storage basins. The size and maximum depth of pits in aluminum alloys are generally not predictable with any accuracy based on analytical models. Some pits initiate, but never progress deeper than a few mils, while others continue to grow. Tests at SRS in fuel storage basins with aggressive water during 1993 showed that pitting of typical aluminum-clad alloys could progress through the 30-mil clad thickness in less than 2 months [9].

Although the size and shapes of pits vary greatly, the mouth of a pit tends to be roughly circular and the pit cavity is somewhat hemispherical. Pits vary from wide saucer-like depressions to small, deep cylindrical holes [10]. During the corrosion process, the thickness of aluminum oxide formed can be related to the amount of aluminum metal consumed during the process by performing a molar balance on a per unit surface area basis. For a temperature range between 30-50  $^{\circ}\text{C}$ , about 0.41 cm of aluminum metal is consumed for every cm thickness of aluminum oxide formed [11]. This means that about twice the thickness of Bayerite and Boehmite aluminum oxide is formed as aluminum metal is consumed. In addition, an acidic solution of aluminum salts is formed by reactions with impurities like sulfates and chlorides in the pit. In the pitting process, this corrosion product is pushed up from the cavity to form nodules on the surface or may fall away into the water leaving the pit exposed.

Corrosion of the uranium-alloy fuel meat is similar to the corrosion of aluminum, Cesium and other fission products are generally associated with the dispersed  $\text{UAl}_x$  particles throughout the aluminum matrix. Release of these fission products by corrosion to the basin water is low.

### 3.3 Visual Examination and Acceptance Criteria

#### 3.3.1 Requirements for Visual Examination

Each fuel assembly to be sent to SRS from FRR shall be examined by a visual test at the storage basin prior to loading for shipment to SRS to determine the condition of the cladding. Binoculars or video imaging equipment may be used as aides for the examination.

The regions of the fuel assembly to be examined are those directly observable through basin water or through air, in the case of dry storage.

Section 3.3.2 provides the acceptance criteria for the examination.

Examination of the fuel condition upon receipt at SRS basins is not required because no additional degradation during shipment is expected.

### 3.3.2 Acceptance Criteria

- 1) No exposure of the fuel meat as a result of i) mechanical damage or cutting, ii) through-clad cracks, or iii) localized corrosion or pitting; and
- 2) No corrosion nodules on the cladding over the fuel meat region greater than approximately 1/8" in diameter.

Fuels whose condition meets the criteria would have a low potential of having a through-clad penetration which could result in release of fuel or fission product species from the fuel meat during shipping to and storage in the Savannah River Site (SRS) basins. These fuels are acceptable for direct storage in SRS basins. Fuels whose condition does not meet the criteria would have to be encapsulated prior to storage in SRS basins.

### 3.4 Basis for Examination Region

The external surfaces of the rectangular shaped MTR fuel assembly are readily visible for inspection as the assembly is brought to within a few feet of the water surface. Only the top edges of the internal fuel plates are visible from above the surface of the water. The two external fuel plates are the most visible and important surface areas of the visual inspection (see Figure 1). These two surfaces are considered to be a conservative representation of the condition of the non-visible surfaces of the fuel assembly because:

- a) The external plates, which surround the interior fuel plates, are more likely to have scratches in the protective oxide coating formed during irradiation of the fuel element, resulting in a higher probability of pitting corrosion than on the interior fuel plates,
- b) Impurities in the water which can plate out or deposit on the aluminum to form localized anodic/cathodic sites have direct access to the external plates, whereas flow to the interior plates is likely lower and somewhat restricted by the geometry.
- c) The external plates are usually the contact points for a galvanic couple between stainless steel or other dissimilar-metal storage racks with corrosion currents highest at these contact locations. Corrosion currents are lower on the interior plates, not only because they are further away from the contact points, but also because the current does not readily flow around corners [12].
- d) Field experience and analysis on un-irradiated MTR type fuel stored in stainless steel storage racks at the IPEN IEA-R1 Research Reactor in Brazil has shown pitting corrosion only on external fuel plates [2]. Examination of Mk 31A slug assemblies stored in SRS L-Basin showed pitting in scratches only on the outer target slugs. The inner slugs remained free of scratches and pitting corrosion [13].

Corrosion products located outside the fuel meat region of the assembly (side plates, end fittings and extremities of the fuel plates) have no potential for the release of fission products, because these locations contain no active fuel meat.

### 3.5 Basis for Examination of Corrosion Nodules

The visual criteria for corrosion nodules on spent nuclear fuel assemblies has been formulated based on four years of corrosion surveillance testing of aluminum alloys in SRS basins and on the analysis of the underwater video of the MTR type aluminum-clad spent fuel at the IPEN-IEA-R1 Research Reactor in Sao Paulo, Brazil [2]. Field inspection was carried out visually using binoculars and up close with an underwater video camera system consisting of an 8mm camcorder and video probe. A total of 66 assemblies were inspected giving a large number of visual scenarios for evaluation in formulating the criteria. Results of this inspection have been reported in Reference 2.

Pitting corrosion on the aluminum-clad spent fuel plates is usually manifested by nodules of white aluminum oxide corrosion product which are readily visible on the external surfaces of the fuel assembly. These nodules may be hard and crusted and range in diameter from about 1/32 inch to 1/4 inch or greater. A 1/4 inch diameter nodule, in most cases, is about 1/4 inch high. In every case, the nodules have pits, or cavities, beneath them as evidenced in the Brazilian fuel inspection. The pits varied in depths, but based on the underwater photography, pits of a diameter of about 1/8 inch, or greater, were seen to be through the 15-mil (0.015 in) clad as judged from comparisons of drilled holes of a known diameter on the edges of the side plates. Nodules of less than 1/8 inch in diameter did not appear to have underlying pits penetrating the cladding.

### 3.6 Release of Species from Exposed Fuel Meat into Water

An evaluation of the release rate of fuel and fission product species has been performed by reviewing literature information and recent field test data on fission product leakage from U-Al alloy fuel material using a sip test.

#### 3.6.1 Sip Tests of FRR SNF

As part of the return of FRR to the SRS, measurements of radioactivity releases have been made by performing sip tests. The tests were made on fuels from European [14, 15] and South American research reactors [2, 16]. The tests in the European countries were made with fuels loaded into LWT shipping casks. In all cases, the results showed that the difference in Cs-137 activity in the cask water before and after a 4-hour minimum rest time was no greater than about 10 pCi/mL which corresponds to a release rate result of 0.9  $\mu$ Ci/hr into a 100 gallon cask.

A controlled field experiment to measure of the cesium release rates from fuels with and without through-clad pits, before and after removal of some of the nodular products were made during field inspections in Brazil [2]. These tests were performed on individual fuel assemblies using special tube containers. The complete set of sip test results from both before and after cleaning (see Table 2 in reference 2) were compared to the acceptable release rate for direct basin storage in SRS basins provided in interim criteria for Cs-137 release rates [3, 4]. Assembly IEA-53, which had the greatest extent of pitting corrosion over the fuel meat (see Figures 1 and 2 in reference 2), released approximately 0.33 pCi/mL/hr or 9.24 nCi/hr into the 28,000 mL of water used in the sip test. A full shipment of 42 of assemblies, even if each released radioactivity at this bounding rate, would yield a rate of 0.39  $\mu$ Ci/hr, well below the interim SRS facility-based criteria of 13  $\mu$ Ci/hr [3, 4].

In principle, the greater the level of exposed fuel meat, the higher the release rate of cesium. In field practice, several conditions make impractical the development of a sip test method to determine precisely whether a fuel is breached or not. These include:

- the low release rates from breached aluminum SNF at basin conditions; and
- surface contamination of the fuel assemblies and shipping casks

The low release rates from breached aluminum SNF are discussed in section 3.6.2. The presence of contamination confounds the measurement of the cesium from any exposed fuel meat. Experience in measuring radioactivity levels in water resting in empty casks (without fuel) has yielded the following results [16, 17]:

Cask type	1st sample	2nd sample	Rest Time	Volume
CD#4 [17]	13 dpm/mL	34 dpm/mL	4 hrs	460 gal
LWT [16]	149 dpm/mL	221 dpm/mL	5.75 hrs	110 gal

Simple calculations from this data show that an empty CD #4 leached approximately 4.65  $\mu\text{Ci/hr}$ , and an empty LWT leached approximately 2.34  $\mu\text{Ci/hr}$  of Cs-137 into the cask water. These rates are a significant fraction of the facility-based limit of 13  $\mu\text{Ci/hr}$  [3, 4].

### 3.6.2 Corrosion and Release Rates of U-Al Fuel

Literature results on the corrosion of U-Al alloys with uranium from 16 to 45 wt% show a slight increase in weight gain compared to 2S aluminum (1100 Al) exposed to water at temperatures of 300°F [18]. The maximum weight gain of the U-Al in these tests was twice that of aluminum. These results are consistent with recent SRS laboratory data showing 10%U-Al and 8001 Al to have a corrosion rate of 0.17 mpy in basin water at room temperature.

A measurement of radioactivity release using sip tests on spent nuclear fuel that was cut to expose the core showed a per-unit-area rate of about 35  $\mu\text{Ci/hr/cm}^2$  [19]. The temperature of the fuel during these tests was not reported except that the fuel was "freshly stored aluminum-uranium alloy fuel elements from the Brookhaven Graphite Research Reactor."

A correlation between the release rate results and the level of breach for aluminum SNF (i.e., release model) is dependent on several factors:

- environment (temperature and quality of the water);
- radioisotope content of fuel (enrichment, burn-up, and decay time);
- fuel material; and
- clad material.

A simple model to estimate the release is based on using a linear corrosion rate for exposed fuel with the assumption that there is no effect from galvanic coupling with the cladding material. A calculation is performed to benchmark this model with the field data. The fuel plates of IEA-53 contained 45% uranium in aluminum, 20% enriched. The plates had a moderate burn-up of 16.7% prior to removal from the reactor in 1962. The present Cs-137

activity of the plates is 1.92 Ci/plate [20]. This corresponds to a fuel core activity of 0.0974 Ci/cm<sup>3</sup>. Using an estimate of exposed fuel area of 10 pits of 1/4 inch (0.635 cm) diameter yields a corrosion rate of 0.1 mil per year. The per-unit-area release rate from assembly IEA-53 is 0.0029  $\mu$ Ci/hr/cm<sup>2</sup>.

The expected release rates to basin water environments for fuels that have breaches in the cladding or exposed fuel meat, are greatly below the existing capacities in basin handling and storage operations at SRS. Under basin conditions, the release rates are so low, even with large breaches of the cladding, that measurement of radioactivity levels above background is difficult.

The radioactivity in the SRS basins is contained in the basin water. The acceptable release rate for the fuels [3, 4] is tied into the facility limit for radioactivity of the water and the ability of the basin deionization system to maintain control. Because the release rates from aluminum SNF are so low at basin storage conditions, cladding breaches pose negligible consequences to worker health or operational constraints.

Due to environmental effects, the release rate of non-volatile radioactive species in air would be bounded by that of water.

The mechanism for the potential release of radioactivity is quite different for the U-Al alloy fuel vis-à-vis commercial reactor spent fuels. In the case of commercial reactor spent fuel rods, a pin-hole breach in the zircalloy cladding essentially provides a direct path for instantaneous release of fission products from the fuel core region to the surroundings. Due to the fuel material and structure of an aluminum SNF element, a cladding breach results in a very low release rate of radioactivity that is barely detectable, especially under basin storage conditions. The acceptance criteria for the direct basin storage of the MTR type aluminum spent fuel herein is thus very conservative with respect to commercial reactor fuels and potential radioactivity release.

### 3.6.3 Estimate of Release from Exposed Fuel at Visual Examination Criteria

Using the sip results from assembly IEA-53, the visually-worst fuel in Brazil (see Figures 1 and 2 of reference 2), an estimate is made of the release of Cs-137 that would result if a 1/8" diameter pit has breached the cladding. Assembly IEA-53 was estimated to contain 10 pits of 1/4 inch diameter. From a simple ratio of the area of 1 pit of 1/8 inch diameter to 10 pits of 1/4 inch diameter, the release rate from a 1/8 inch diameter pit would be  $(0.0123/0.491) \times 0.33 \text{ pCi/mL/hr} \times 28,000 \text{ mL} = 231 \text{ pCi/hr}$  (0.000231  $\mu$ Ci/hr). This rate of release of activity is not practically detectable unless long rest times (several days) and/or high resolution and sensitive detection systems are used.

## 4.0 IMPLEMENTATION OF VISUAL EXAMINATION CRITERIA

Figure 1 shows a schematic of an MTR assembly with regions of postulated corrosion degradation. On the external fuel plates, the fuel meat region is usually distinguishable from the outer edges of the plate by a slightly darker area (Figure 1). Outside this region, the plate is aluminum. Where this plate fits into the grooves of the two side plates (Figure 1), a crevice is formed. White nodular corrosion product in this area outside the fuel meat can form as crevice corrosion, but is not detrimental as long as it is in the crevice and outside the fuel meat region. The designed dimensions of MTR assemblies show that the

fuel meat extends to within 1/32 to 1/16 inch of the side plate and at least 1 inch from the top and bottom of these fuel plates. Observations [2] have shown that as much as 1/4 inch of non-fuel meat region is visible, in some cases, on the edges of outside fuel plates. Based on the size of corrosion nodules and the location of these nodules, the examiner can judge if the crevice corrosion has caused a potential breach in the fuel meat region. Corrosion nodules outside the fuel meat will have no impact on fission product release or structural integrity. Corrosion nodules of approximately 1/8 inch diameter directly over or extending into the fuel meat region are unacceptable.

## 5.0 ADDITIONAL RECOMMENDATIONS

Fuels that do not meet the acceptance criteria may be encapsulated. As an alternative to encapsulation for interim storage, direct storage of minor breached aluminum SNF is technically feasible. Release of radioactivity to the SRS basin water is a contained release. Removal of radioactive species and non-radioactive contaminants through deionization is part of the routine operation of the basins. Storage of breached assemblies directly into basin water without encapsulation would be acceptable if the release rates are within those of deionization makeup, ensuring that facility health and safety limits are not violated.

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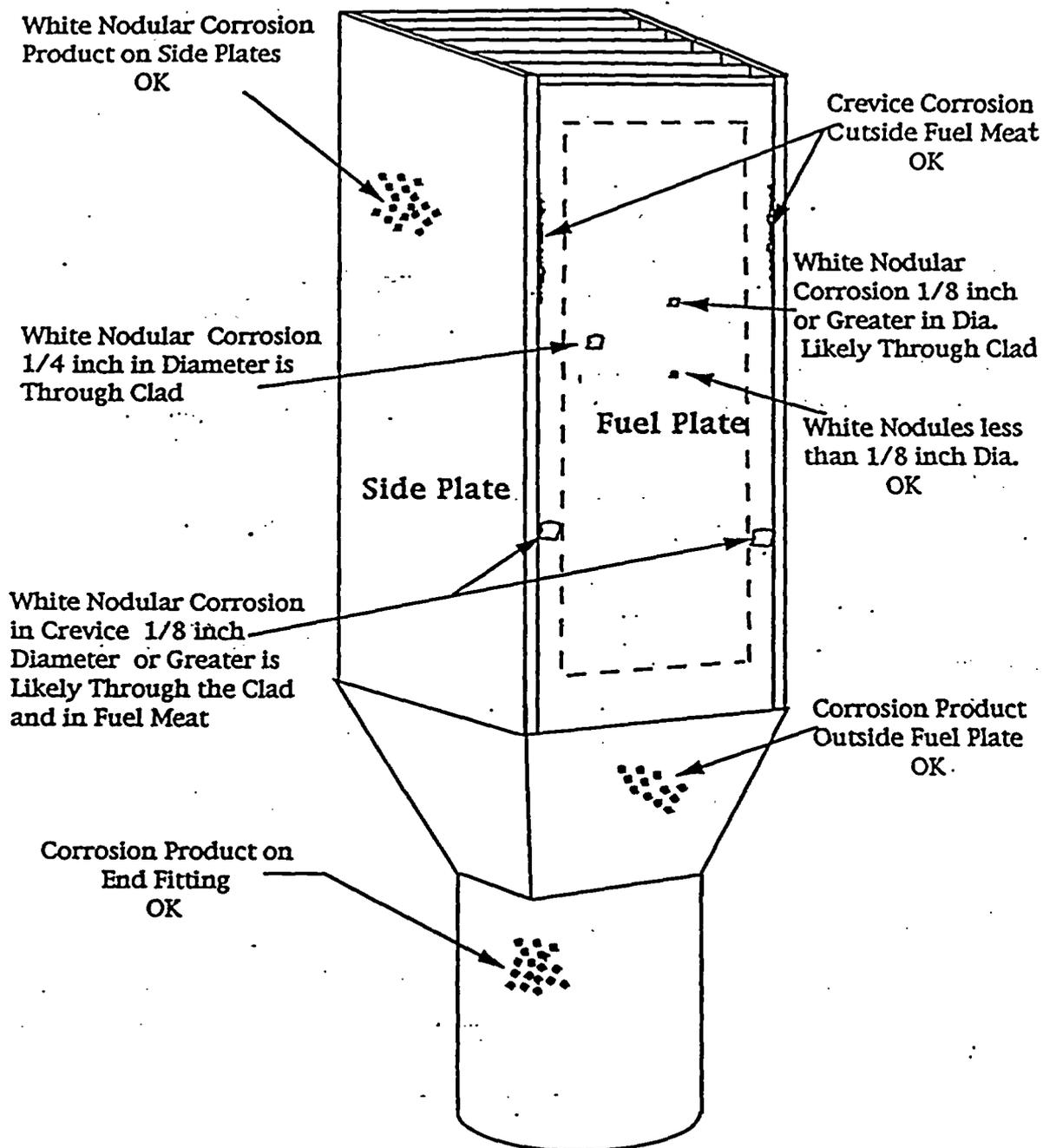


Figure 1 - Schematic of MTR Assembly with Examples of Pitting and Crevice Corrosion

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# Radioactivity Release from Aluminum-Based Spent Nuclear Fuel in Basin Storage (U)

R. L. Sindelar and J. P. Howell

Savannah River Technology Center  
Strategic Materials Technology Department  
Materials Technology Section

Publication Date: May 1997

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Savannah River Site  
Aiken, SC 29808**

This document was prepared in connection with work done under Contract No. DE-AC09-96SR18500 with the U. S. Department of Energy

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**DOCUMENT:** WSRC-TR-97-0153

**TITLE:** Radioactivity Release from Aluminum-Based Spent Nuclear Fuel in Basin Storage (U)

**APPROVALS**

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\_\_\_\_\_  
R.L. Sindelar, Author  
Materials Applications & Corrosion Technology Group  
MATERIALS TECHNOLOGY SECTION

Date: 5-30-97

  
\_\_\_\_\_  
J. P. Howell, Co-author  
MATERIALS TECHNOLOGY SECTION

Date: 5-30-97

  
\_\_\_\_\_  
M. R. Louthan, Jr, Technical Reviewer  
MATERIALS TECHNOLOGY SECTION

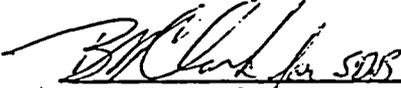
Date: 5/30/97

  
\_\_\_\_\_  
N. C. Iyer, SRTC-Spent Nuclear Fuel Manager  
Materials Applications & Corrosion Technology Group  
MATERIALS TECHNOLOGY SECTION

Date: 5/30/97

  
\_\_\_\_\_  
T. L. Capeletti, Manager  
MATERIALS TECHNOLOGY SECTION

Date: 5/30/97

  
\_\_\_\_\_  
S. D. Burke, Manager  
SPENT FUEL STORAGE DIVISION

Date: 5/30/97

**SUMMARY**

This report provides an evaluation of: 1) the release rate of radionuclides through minor cladding penetrations (breaches) on aluminum-based spent nuclear fuel (Al SNF), and 2) the consequences of direct storage of breached Al SNF relative to the authorization basis for SRS basin operation. The breached reference fuel assembly (BRFA), a fuel assembly at a conservative upper bound reference condition, is defined by a:

- Cs-137 loading of 73.1 Ci/in<sup>3</sup> of fuel material<sup>1</sup>; and an
- Exposed fuel core region of 0.5 in<sup>2</sup>.

A model for the release rate from breached fuel in water is developed from consideration of the materials and structure of Al SNF. The model was validated using field data on an assembly with cladding penetrations totaling 0.5 in<sup>2</sup> of exposed fuel core which bounds the most severely breached fuel assemblies expected to be received in the Foreign Research Reactor Fuel program. A release rate of 9.24 nCi/hr of Cs-137 was measured for this fuel assembly and is in good agreement with the rate of 15.5 nCi/hr conservatively predicted from the release rate model. Using the release model, the estimated release rate from the BRFA into basin water is 0.835 µCi/hr (835 nCi/hr) of Cs-137.

The consequences of storing breached fuel relative to the authorization basis is evaluated through the postulated storage of 1500 BRFAs, a number approximately 10% of the expected receipts from the Foreign Research Reactor program over the next 5 - 10 years. Calculations, based on this storage scenario for both the L-Basin and the Receiving Basin for Off-site Fuels (RBOF), show that the basin water activity levels increase, but remain well within the authorized limits (see Table 1).

**Table 1- Estimated Changes in Activity Levels in the SRS Basins for Storage of 1500 Assemblies Each Releasing 0.835 µCi/hr of Cs-137**

Basin	Present Steady-State Basin Activity Level w/Deionization (dpm/ml)	Estimated Change in Steady-State Basin Activity Level w/Deionization* (dpm/ml)	Present Activity Increase Rate w/Inactive Deionization (dpm/ml/day)	Estimated Change in Activity Increase Rate w/Inactive Deionization* (dpm/ml/day)	Authorized Limit for Water Activity (dpm/ml)
L-Basin	4	61.1	0.31	5.22	2000 (Cs-137)
RBOF	259	123	36	35.2	220,000 (total)

\*Estimated contribution from the postulated storage of the 1500 BRFAs only

The present activity increase in the basins when the deionization systems are inactive is approximately 0.31 dpm/ml/day for L-Basin [1] and 36 dpm/ml/day for RBOF [2]. The current source term for Cs-137 in the L-Basin includes releases from the inventory of "at risk" Mk-16 assemblies together with releases from the basin walls, sludge, and fuel

<sup>1</sup>The total cesium content of the BRFA is equal to that of the Reference Fuel Assembly (RFA) used in the authorization basis for the Receiving Basin for Offsite Fuels (RBOF)

surface contamination. The RBOF source term includes releases from the inventory of failed Taiwanese Research Reactor fuel, Heavy Water Components Test Reactor fuel and several other degraded fuels. These rates would increase with the storage of 1500 BRFAs to totals of 5.53 and 71.2 dpm/ml/day, respectively for L-Basin and RBOF. In the event of inactive deionizer operation when storing 1500 BRFAs, the authorized Cs-137 activity level of 2000 dpm/ml for L-basin [3] and total activity of 220,000 dpm/ml for RBOF [4], would not be exceeded for 350 days and 8.5 years, respectively. This provides sufficient time to implement portable deionizer systems that have successfully been used in the past, if such implementation was necessary.

The acceptance criteria for the physical condition of Al SNF for direct storage in SRS basins [5] is derived from present requirements for receipt of foreign research reactor (FRR) fuel [6] and excludes fuels with cladding breaches without canning. This present evaluation predicts the effect of direct storage, without canning, of Al SNF with minor cladding penetrations on basin activity. These results demonstrate that operations would remain well within the authorization basis.

## INTRODUCTION

Approximately 20 metric tons or approximately 15,000 aluminum-based spent nuclear fuel (Al SNF) assemblies will be shipped from foreign research reactors (FRR) to the Savannah River Site (SRS) over the next 10-15 years. These assemblies will be stored at SRS in the L-Basin and the RBOF. A number of these fuel assemblies have corrosion or mechanical damage that has resulted in minor penetrations (breaches) of the aluminum cladding and exposure of the fuel core to the basin water. Corrosion of this exposed core will cause release of radionuclides to the basin water. This report provides an evaluation of the release rates of radionuclides from breached Al SNF assemblies and evaluates the effect of direct storage of breached fuel at a conservative upper bound reference condition on the basin water activity level and waste generation.

## DISCUSSION

### Aluminum-Clad Spent Nuclear Fuel

Aluminum-based spent nuclear fuel (Al SNF) is made by powder metallurgy or by casting and rolling. The fuel core for most of these fuels is an aluminum matrix in which particles (dispersoids) of either  $UAl_x$ ,  $U_3Si_2$ , or  $U_3O_8$  oxide particles are embedded. The core is protected by a cladding of aluminum alloy which is metallurgically bonded to the core.

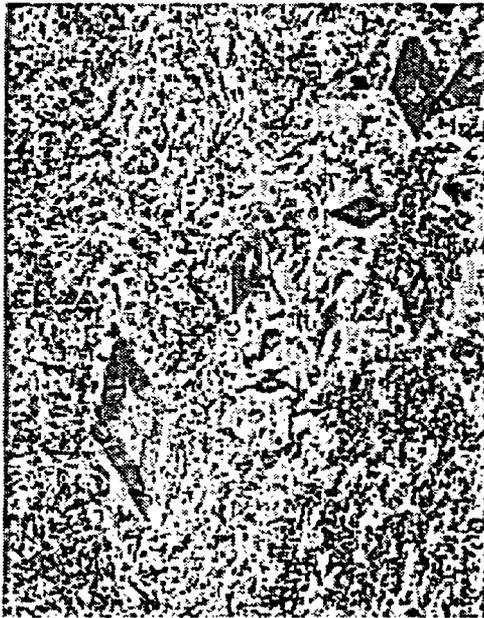
The general structure of the fuel core material, dispersoids in aluminum (Figure 1), is retained following irradiation [7]. Small, uniformly-distributed pockets of fission product gases form within the fuel during irradiation for the  $UAl_x$  and  $U_3O_8$  dispersoid fuels; the largest pockets of gas are formed in the high-burnup  $U_3Si_2$  dispersoid fuel (Figure 2). The gas pockets are not interconnected. Otherwise, no significant redistribution of the fission products occurs. That is, the fission products are uniformly distributed throughout the fuel core, the fuel/cladding bond is retained, and if the cladding becomes penetrated, only the immediate fuel core area at the point of breach is exposed.

The mechanism for the potential release of radioactive species is quite different for the Al SNF fuel than for commercial reactor spent fuels. In the case of commercial reactor spent fuel rods, a pin-hole breach in the zircalloy cladding essentially provides a direct path for

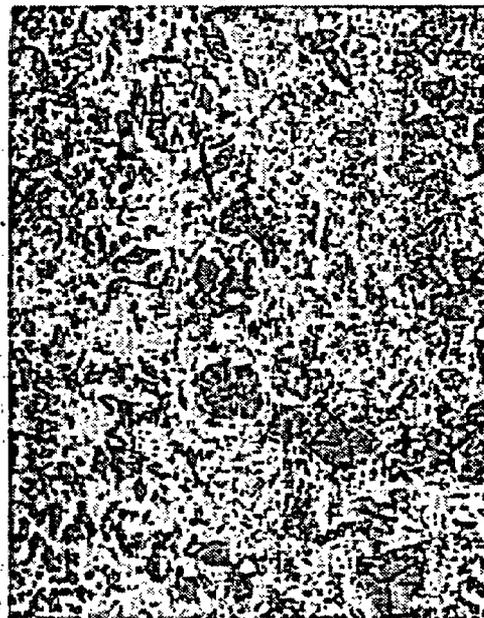
release of fission products from the fuel core region to the surroundings. This is not the case in Al SNF because the fission products are contained locally in the fuel core region.

Breaches in the claddings of Al SNF typically occur through pitting corrosion or mechanical damage. Pitting corrosion is prevalent only under aggressive water chemistry conditions [8-10]. Appendix 1 provides an additional discussion on the corrosion of Al SNF in basin storage.

Although there are multiple types of aluminum-based fuel, all have the common structure of dispersoids in aluminum and are expected to have similar corrosion behavior and release behavior in high purity water at basin temperatures. The corrosion rate of the exposed fuel decreases slowly with time as a passive film grows on the fuel material. A cladding breach results in a very slow release rate of radioactivity under the low conductivity, high purity storage conditions of the SRS basins.



(a) Aluminum-18 wt% Uranium  
Longitudinal Direction (500X)



(b) Aluminum-33 wt% Uranium  
Longitudinal Direction (500X)

**Figure 1- Microstructure of U-Al Alloy Fuel Core Material**

### Model for Radioactivity Release Rate from Al SNF

A model for the radioactivity release rate of fuel with cladding breaches is needed for estimating the release rates from Al SNF representative of the expected receipts. In general, the release rate of radioactivity from Al SNF for a given level of cladding breach is dependent on several factors:

- environment (temperature and quality of the water);
- radioisotope content of fuel (enrichment, burn-up, and decay time);
- fuel core material (post-irradiation composition and microstructure); and
- clad material

At the low temperatures typical of basin storage, corrosion is the primary mechanism whereby species from the fuel core are released into the water. The radionuclides are conservatively assumed to be not bound in the corrosion product, but rather are fully soluble and free to disperse into the water. No significant galvanic interaction between an aluminum-uranium fuel material and aluminum cladding has been measured [10]. Therefore, a simple model to estimate the release from fuel core is developed by considering general corrosion of the fuel core region directly exposed to the environment. The release model is given by:

$$R = A \times B \times C$$

where R [Ci/hr] is the total radioactivity release rate;

A [Ci/in<sup>3</sup>] is the total radioactivity in the fuel core material at the decay time of interest;

B [in<sup>2</sup>] is the area of fuel exposed to the environment (area of breach); and

C [in/hr] is the general corrosion rate of the fuel core material in the environment of exposure (i.e. the corrosion rate is a function of temperature and water quality).

This model can be applied to water storage at temperatures where general corrosion of the exposed fuel is the primary release mechanism (< 200°C). Above this temperature, i) hydrogen blistering attack and exfoliation of the fuel material could occur greatly increasing the rate of attack of the fuel material; and ii) diffusion of radioactive species in the fuel material could occur to allow communication of the bulk fuel core with the exposed surface in direct contact with the water.

Results from laboratory corrosion tests at the Savannah River Technology Center show that both 10%U-Al and 8001 aluminum cladding with fresh surface finishes (600 grit) have a corrosion rate of 0.17 mils per year ( $2 \times 10^{-8}$  in/hr) in basin water at room temperature. The fresh surface corrosion rate is expected to be similar for all aluminum-based fuel core materials and can be used as an upper bound estimate of the corrosion rate in basin water. The corrosion rate should decrease slowly with time as a passive film grows on the fuel material in a manner similar to the corrosion of aluminum in water at temperatures less than 100°C [11]. For the purpose of this study, a corrosion rate of 0.2 mpy is used to bound the corrosion rate of exposed Al SNF fuel core material in basin water.

### Measurements of Release of Radioactivity from Al SNF

Radioactivity releases from spent fuel can be measured directly by performing a "sip" test. The test is performed by measuring the activity of Cs-137 in a specified volume of water before and after the fuel "rests" in the water for a specified period. A special field

experiment using this testing method to measure the cesium release rates from individual fuel assemblies was performed with Materials Test Reactor (MTR) assemblies in Brazil [12]. This experiment involved using fuels with and without through-clad pits, as well as, fuels before and after removal of some of the surface corrosion nodules. A summary of the field examinations is contained in Appendix 2 of this report. Assembly IEA-53, having the greatest extent of pitting corrosion over the fuel meat (see Appendix 2), released 9.24 nCi/hr at a constant rate [5, 12].

A validation of the release model discussed above is performed by the following calculation using the data from assembly IEA-53. The fuel plates of IEA-53 contained 45% uranium in aluminum, 20% enriched in U-235. The plates had a moderate burn-up of 16.7% prior to removal from the reactor in 1962. The present Cs-137 activity of the plates is 1.92 Ci/plate [13]. This corresponds to a fuel core activity of 1.36 Ci/in<sup>3</sup>. Using an estimate of exposed fuel area of 10 pits of 1/4 inch (0.635 cm) diameter exposes 0.5 in<sup>2</sup> (3.2 cm<sup>2</sup>) of fuel core material and a bounding corrosion rate of 0.2 mils per year, the predicted release rate is 15.5 nCi/hr. This result is in conservative agreement to the measured release rate of 9.24 nCi/hr.

The release of radionuclides is due to aqueous corrosion. Removal of breached Al SNF from water and exposure to air will stop this corrosion, and mitigate the release of radionuclides. Measurements of radioactivity release for fuels heated in air are being performed under a separate Savannah River Technology Center program conducted at Argonne National Laboratory. The results of recent tests to measure the species released from exposed fuel heated at 275°C in air have shown no detectable release of any radionuclides from U<sub>2</sub>Si<sub>3</sub>-Al fuel in a one-month test [14]. The microstructure of the fuel specimen used in this test is shown in Figure 2.

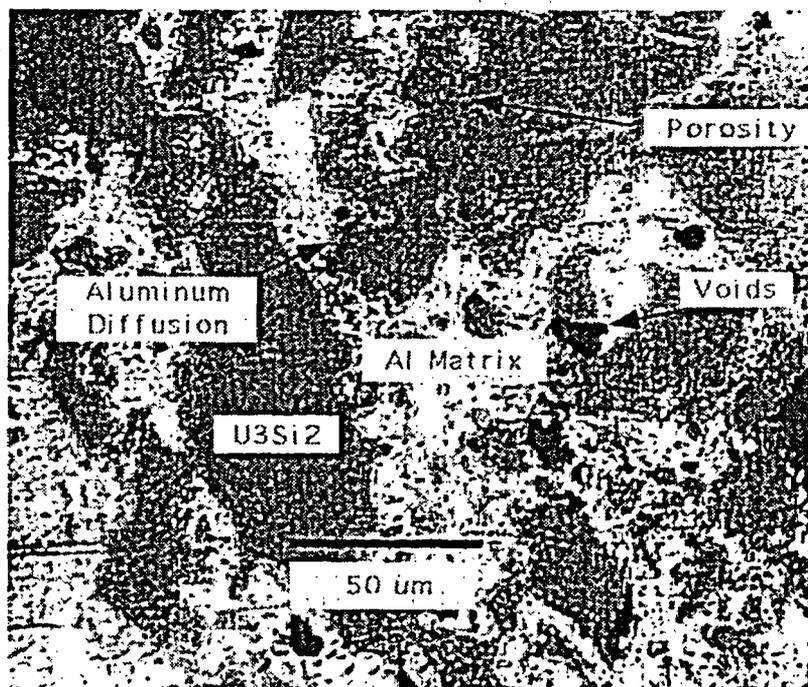


Figure 2- Microstructure of U<sub>2</sub>Si<sub>3</sub>-Al Fuel Core following 51.4% Burn-up (500X) [14]. The black regions are the internal gas pockets

## Impact of Storage of Breached AI SNF on the Authorization Basis for the Operation of the SRS Basins

### Consequences of Release of Activity from the SRS Basins

The authorization basis for L-Basin and for RBOF are set forth in their respective Basis for Interim Operation [3, 4]. The safety envelope is established by analyzing the risks associated with various accident scenarios and is based on specific assumptions regarding basin operations, including basin water radionuclide content. The authorization basis for L Basin [3] limits the basin water cesium concentration to 2000 dpm/ml. Under a worst case accident scenario during which the entire 3.5 million gallons of water at 2000 dpm/ml is instantaneously evaporated and released to the environment, the consequence to the public is a total dose of 16 mR. The on-site worker is exposed to dose rate of 14 mR/hr, within DOE radiological working limits.

The RBOF authorization basis assumes a total basin water activity concentration of 220,000 dpm/ml (0.1  $\mu$ Ci). The concentration values are higher than the L-Basin values because of the smaller volume of basin water, 500,000 gallons, and past operating history. The maximum activity of the basin water in RBOF has been 220,000 dpm/ml in one of the isolated basins during fuel handling and cutting operations [4]. A total release of the RBOF water inventory is negligible because it is bounded by an accident analysis of the estimated exposure from a hypothetical tank farm release. The tank farm release assumes a radionuclide inventory release significantly higher than the RBOF water inventory and consequences of worst case scenario for RBOF, similar to L-Basin, is negligible. Therefore, dose estimate impacts to the public and on-site workers from the operation of RBOF were not estimated in its authorization basis.<sup>2</sup>

The basin facilities are managed and operated to much lower limits of radionuclide concentrations through deionization to further improve the safety of facility, maximize ALARA, and minimize waste generation. Although process requirements limit the L Area cesium activity limit to 1000 dpm/ml, the current steady-state cesium activity level is less than 4 dpm/ml [1]. The current steady-state RBOF activity level is 259 dpm/ml [2] compared with 15,500 dpm/ml total activity, the Technical Safety Requirement for the facility.

Other radionuclides potentially released from breached fuels include gaseous fission products and noble gases. The risks associated with the release of these radionuclides are bounded by the accident scenario involving an inadvertent criticality in the basins. The postulated criticality releases significantly more gaseous fission products than that released from a breach in the fuel cladding. The doses to the public associated with an inadvertent criticality are insignificant relative to other risks associated with the the L-Basin and relative to other RBOF risks.

The activity release into the basin water is based on the potential releases associated with damaging a reference fuel assembly (RFA) [3]. The release of gaseous fission products and noble gases are not contained in the basin water. In accordance with Safety Guide 1.25 [15], damage of fuel results in the release of 30% of the Kr-85 and 10% of other

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<sup>2</sup>A Safety Analysis Report (WSRC-SA-11) for RBOF is undergoing final approval. The results in this present report are consistent with the SAR.

and noble gases are not contained in the basin water. In accordance with Safety Guide 1.25 [15], damage of fuel results in the release of 30% of the Kr-85 and 10% of other noble gases and iodine. A bounding analysis conservatively assumes that 2200 RFA's are damaged concurrently, each containing 1050 curies of Kr-85. No other noble gases are present in the isotopic inventory (see Table 6-1 of reference 3 of this report) as a result of the two year cooling period. A comparison of the total radiological release to the minimum radiological source terms required to meet consequence level thresholds (Tables 6-10 and 6-11 of reference 3) results in a determination of low consequences to the facility worker (< 25 rem), the onsite worker (< 500 mrem), and the maximum exposed offsite individual (public, <100 mrem).

Another important factor in the operations of the basins is the consequence of the cesium loading of the ion exchange resin beds of the deionizer system. The L Area deionizer system design basis was an 8 curie loading of the zeolite beds used for cesium removal. Shielding was designed to maintain the worker dose less than 1 mR/hr. Currently, the zeolite beds only retain 2 curies of cesium before replacement is required because of accumulation of other dissolved solids in the mixed beds. The postulated addition of 1500 breached clad assemblies would increase the zeolite loading from 2 curies to slightly less than 8 curies, before change out, still within the design basis of the system, safety authorization, and with no increase in waste volume. Similarly, the RBOF resin beds are regenerated twice per year, and no change to the schedule or additional waste volume would result due to the storage of 1500 BRFAs. Only the curie loading would increase. Shielded cells and remote resin handling equipment preclude additional worker dose.

#### Impact of Postulated Release Rates on Water Activity Relative to the Authorization Basis

As stated, the Basis for Interim Operation of the SRS basins assumes a limit of 2000 dpm/ml for Cs-137 for L-Basin and 220,000 dpm/ml total activity for RBOF [3, 4]. The steady-state activity level during basin operation is dependent on the deionization capacity and the releases into the basin from sludge, basin walls, contamination on spent fuel, and exposed fuel material. With the new deionization system in L-Basin, the current water activity level is less than 4 dpm/ml [1]. The current water activity level in RBOF is 259 dpm/ml [2].

A calculation is performed for the postulated storage scenario of 1500 Al SNF assemblies with breached fuel cladding to evaluate the increase in activity level of the water. A breached reference fuel assembly (BRFA) is developed from the Reference Fuel Assembly (RFA) used in the safety basis for the SRS basins [3] and which bounds the worst case breach observed in the field inspections performed through the Foreign Research Reactor Fuel receipts. The Cs-137 loading of the RFA is 9280 Ci [3]. The RFA is actually for a storage location rather than an individual assembly and six assemblies are typically stored per location. The research reactor fuels are primarily the Materials Test Reactor design. These fuels have 18 fuel plates each with a nominal fuel core volume of 23.5" x 2.5" x 0.02". The volumetric Cs-137 loading of the BRFA is simply calculated to be 9280 Ci per storage location/(6 assemblies per storage location x 18 fuel plates per MTR assembly x 23.5" x 2.5" x 0.02" per plate) or 73.1 Ci/in<sup>3</sup>.

A value of 0.5 in<sup>2</sup> of exposed fuel core is equivalent to ten 1/4" diameter through-clad pits, equivalent to the worst pitted assembly observed during the field inspections to date. Using 1500 BRFAs with 0.5 in<sup>2</sup> of exposed fuel is bounding and takes into account the

possibility that some fuels could have more or less exposed core material. Thus, the BRFA is defined as a reference fuel assembly with a:

- Cs-137 loading of 73.1 Ci/in<sup>3</sup> of fuel material; and an
- Exposed fuel core region of 0.5 in<sup>2</sup>.

Using the release rate model described above, the release rate from the RBFA in basin water is 0.00731 Ci/year, or 0.835  $\mu$ Ci/hr.

Under steady-state conditions, the deionizer system will remove soluble radioactivity (e.g. Cs-137) and suspended insoluble radioactive particles at a rate equal to the total release rate into the basin. The removal rate is a function of the system flow rate and the radioactivity concentration of the water. This steady state condition is expressed as follows:

$$Q \cdot C = A \cdot V$$

where: Q = flow rate through the system (gal/day)

C = steady-state activity of the water (dpm/ml)

A = activity release rate to the water (dpm/ml/day)

V = basin volume (gal)

The deionizer systems at SRS are designed for continuous operation. The average flow rate, Q, for the L-Basin deionizer is assumed to be 200 gpm. The volume of L-Basin is 3,375,000 gallons. For a postulated scenario of 1500 BRFAs, each releasing Cs-137 at 0.835  $\mu$ Ci/hr into the basin, A is calculated to be 5.22 dpm/ml/day. The steady-state concentration C is calculated to be 61.1 dpm/ml. This is the estimated increase to the present steady-state concentration of approximately 4 dpm/ml. Recent trending data collected with the L-Basin deionizer system inactive shows the activity increase rate to be approximately 0.31 dpm/ml/day [1] resulting from the inventory of "at-risk" Mk-16 fuel, basin sludge, and basin wall contamination. The graph in figure 3 shows that the L-basin deionizer system could be inactive for 350 days before the authorized limit would be exceeded. In comparison, the same calculation performed for 1500 assemblies releasing Cs-137 at 9.24 nCi/hr, the release rate measured for assembly IEA-53, results in a release rate of 0.0578 dpm/ml/day and a steady-state concentration, C, of 0.475 dpm/ml; the deionizer system could be inactive for 14.9 years before the authorized limit would be exceeded.

For RBOF, the flow rate through the deionizer, Q, is approximately 100 gpm and the basin volume is 500,000 gallons. For the same postulated scenario of 1500 BRFAs each releasing Cs-137 at 0.835  $\mu$ Ci/hr into the basin, A is calculated to be 35.2 dpm/ml/day. The steady-state concentration C is calculated to be 123 dpm/ml; this is the estimated increase to the present steady-state concentration of 259 dpm/ml [2]. Recent trending data collected with the RBOF deionizer system inactive shows the activity increase rate to be approximately 36 dpm/ml/day [2]. The graph in figure 4 shows that the RBOF system could be inactive for 8.5 years before the authorized limit would be exceeded. This provides sufficient time to implement portable deionizer systems, if necessary.

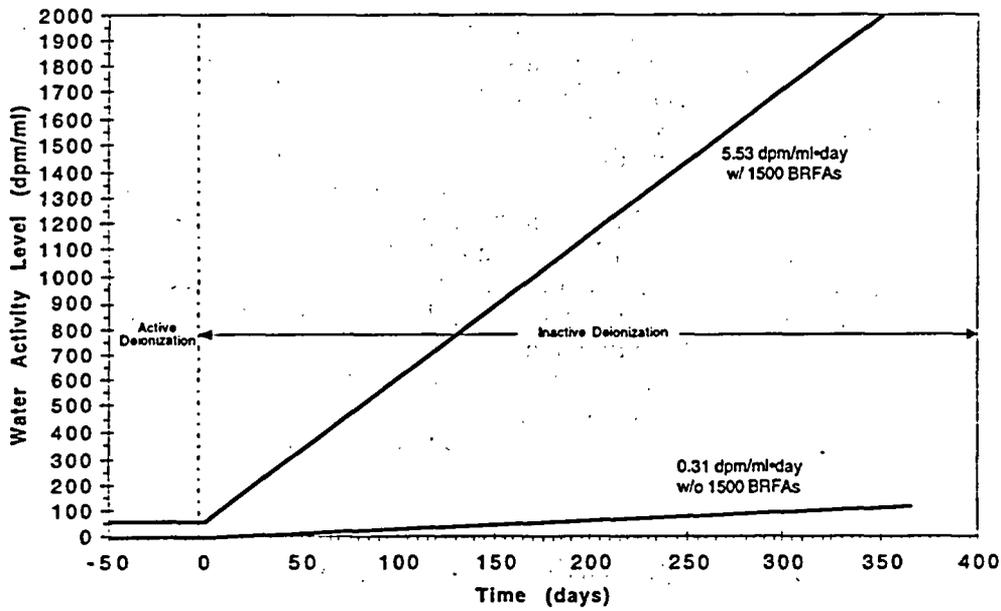


Figure 3- Water Activity Level for L-Basin With and Without Storage of 1500 MTR Assemblies Each Releasing Cs-137 at 0.835  $\mu$ Ci/hr

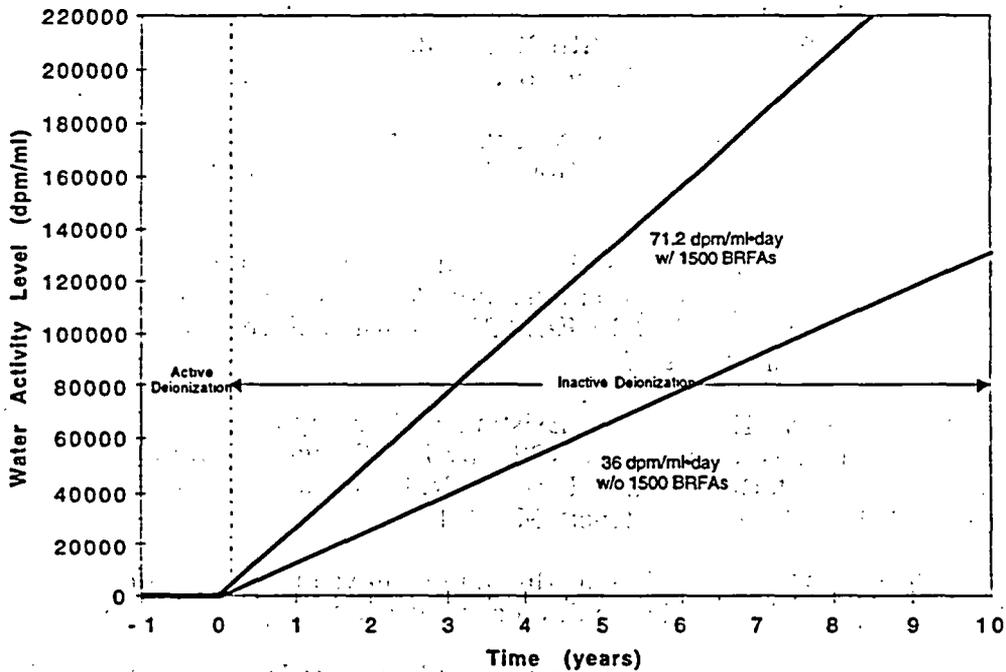


Figure 4- Water Activity Level for RBOF With and Without Storage of 1500 MTR Assemblies Each Releasing Cs-137 at 0.835  $\mu$ Ci/hr

## CONCLUSIONS

A release rate model was developed for aluminum-based spent nuclear fuel (Al SNF) with breached cladding. The measured release rates from fuels in basin storage are consistent with the model. The anticipated release for a conservative bounding scenario of 1500 breached reference fuel assemblies (BRFAs) stored in SRS basins was determined. The application of the model showed that the storage of each BRFA will result in a release rate of 0.835  $\mu\text{Ci/hr}$  of Cs-137. With active deionization, the basin water activity levels are estimated to be 65.1 dpm/ml and 382 dpm/ml, well below the authorization basis limits of 2000 dpm/ml and 220,000 dpm/ml for L-Basin and RBOF, respectively. In the event the deionization systems are inactive, the authorized limits would not be exceeded for 350 days and 8.5 years for L Basin and RBOF, respectively. Fully demonstrated portable deionizers could be brought online within these time periods, if necessary. Although storage of the breached fuels would cause an increase in the curie loading of the waste streams, there would be no increase in the volume of waste generated. The increased curie loading is within the design bases of the facilities and allowed under the authorization bases.

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## APPENDIX 1- Corrosion of Aluminum-Clad SNF in Wet Basin Storage

Corrosion of aluminum alloys and production fuels at SRS and other water storage facilities has been documented [1-3]. Corrosion of the aluminum-clad and the uranium-aluminum alloy fuel core in water storage basins can be attributed to poor quality water at some point in the fuel storage lifetime. Corrosion surveillance tests at SRS have shown that storage in poor quality water can result in pitting corrosion of aluminum alloys. Pit depths of 30 mils, equivalent to the cladding thickness of SRS fuel, were measured on coupons exposed to aggressive basin conditions for 2 months in water with a conductivity of approximately 200  $\mu\text{S}/\text{cm}$ . Under aggressive water condition, the corrosion product on the aluminum cladding is characterized, generally, by crusted nodules of aluminum oxide located primarily in scratches in the protective oxide coating formed during fuel irradiation [4]. These thinned areas make it easier for aggressive ions like chlorides to attack the aluminum. As aluminum oxide is formed, an occluded cell with an aggressive, acidic environment can develop beneath the nodule. This can result in a rapid pit formation in the alloy, penetrating the cladding thickness in short time periods. Visible signs of crusted oxide nodules on aluminum indicate that pitting corrosion has taken place and there is attack beneath these nodules and a high probability of penetration of the cladding.

Pitting corrosion is difficult to predict from water composition alone because of the synergistic nature of the aggressive impurities and the variables involved [1-3]; however, recent laboratory data have shown that water solutions with conductivities less than 50  $\mu\text{S}/\text{cm}$  are non-aggressive to cause pitting corrosion and that the chloride ion may be the controlling factor for pit initiation under basin conditions [5]. The major environmental factors believed to influence the pitting of aluminum alloys are [1-3]: conductivity, pH, bicarbonate, chloride, sulfate, and oxygen content of the water. In addition, the corrosion process is enhanced by the aluminum/steel dissimilar metal couples which exist throughout most basins. Heavy metals like copper and mercury and noble metals like silver can plate out on the aluminum forming strong localized anodic/cathodic sites which promote aggressive pitting of the aluminum.

In contrast, pitting corrosion has not been seen on aluminum cladding and corrosion surveillance coupons stored in high quality water. The water in RBOF has been maintained with a conductivity in the 1-3  $\mu\text{S}/\text{cm}$  range and aggressive impurity concentrations in the parts-per-billion range. After an extensive campaign over the last several years to cleanup the reactor basins which has included sludge removal, new continuous deionization equipment installation, and new aluminum storage racks, all basins at SRS are being maintained below 4  $\mu\text{S}/\text{cm}$ . No new pitting corrosion has been seen in the reactor basins since 1993.

Field tests have shown that pitting corrosion is mitigated when the pitted materials are exposed to high quality water. Fuels that contained through-clad pits with surface corrosion nodules were measured to determine radionuclide release before and after removal of the nodules using a sip test with high purity, low conductivity water [6]. The release rates of Cs-137 from the pitted fuel before and after the removal of the nodules were identical which indicates that an aggressive water chemistry does not exist under the nodules when placed in high quality basin water.

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## APPENDIX 2- Examination of Brazilian Research Reactor MTR Fuel

A team of scientist and engineers from SRS visited the Institute for Nuclear Energy Research of the Nuclear Energy Commission at Sao Paulo (IPEN/CNEN-SP), Brazil July 22-31, 1996 [1] to characterize the extent of corrosion and the effect of this corrosion on the release of radionuclides from this fuel to the research reactor basin water.

Using an underwater video camera system, along with an 8mm video camcorder above the surface of the basin, detailed inspections were made on 66 SNF assemblies which had been stored in the basin water environment for 25-35 years. Nodules of white corrosion products on the outside fuel plates of 41 assemblies were visible to the eye from above the water surface. Hard crusted mounds of aluminum oxide ranged in sizes from about 1/32 inch to 1/4-1/2 inch in diameter as estimated by comparison with drilled holes in the edges of the side plates known to be 0.096 inches in diameter. These oxide nodules are shown on an external fuel plate from IPEN fuel assembly No. 53 below in Figure 1.

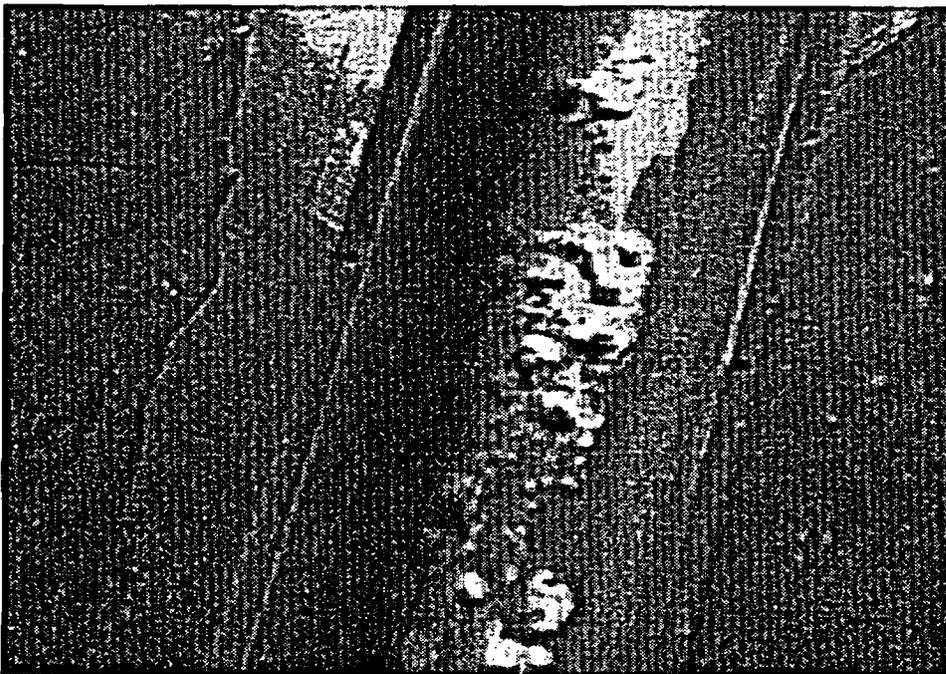
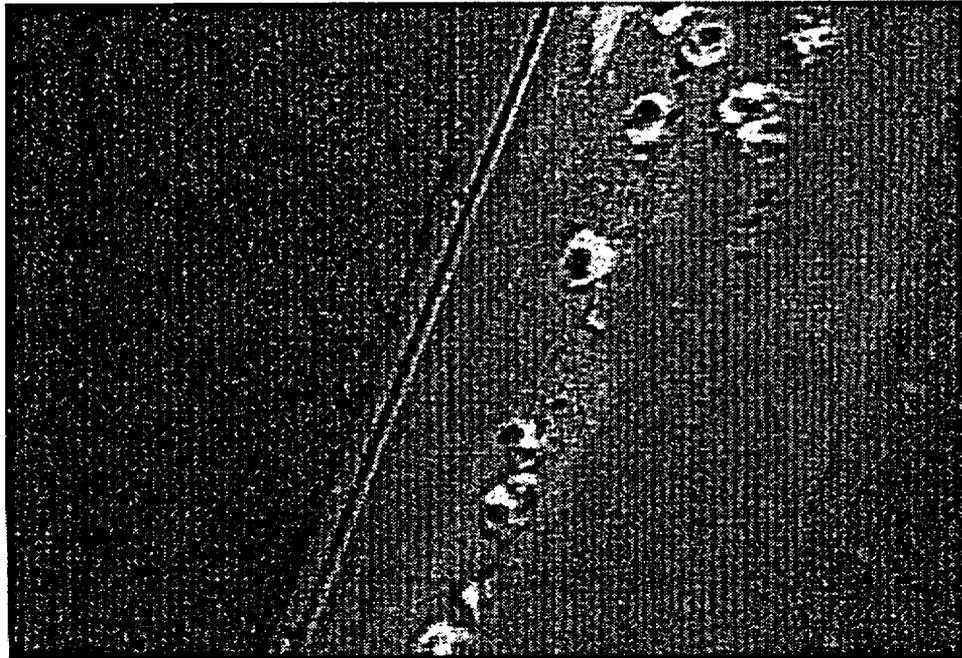


Figure 1- Assembly IEA-53 Before Removal of Nodules

This assembly was selected for an in-depth examination because it showed among the highest leach rates of Cs-137 as determined by a sip test. In addition, this assembly had the largest deposits of visible corrosion product over the fuel plates. This corrosion was typical of the corrosion seen on most of the 41 assemblies. Without exception, every large nodule of oxide of was associated with a thru-clad pit in the aluminum cladding beneath the corrosion product. This was determined by brushing or scraping the oxide nodule to remove from the fuel plate to reveal the pit below. Figure 2 shows typical through-clad pits after oxide removal from some of the nodules on the fuel plate of assembly IEA-53. The pits were judged to be through the 0.015 mil cladding by comparison with the known 0.096 mil diameter holes drilled in the side plates. Analysis of the nodular corrosion product using gamma ray spectroscopy showed uranium-235, europium-154, and cesium-137, confirming that the pit associated with the nodule had breached the cladding into the fuel core.



**Figure 2- Assembly IEA-53 After Removal of Nodules**

At the time of this underwater fuel inspection, this visual examination was the most extensive examination of AI SNF in water storage basin in the world. It showed that based on the size of the visible corrosion nodule as compared to known reference dimensions on the fuel, a judgment can be made on the size of the pit beneath this product. As the cladding thickness on the research reactor fuel is relatively thin compared to the diameter of the oxide nodules, the pits generated in the surface of the fuel plates have a high probability of penetrating the clad into the U-Al core.

The details of the sip tests of the Brazil fuels are contained in reference 1.

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