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SPENT NUCLEAR FUEL SAMPLING STRATEGY

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SPENT NUCLEAR FUEL SAMPLING STRATEGY

1.0 INTRODUCTION

1.1 PURPOSE

The main objective of this report is to provide a strategy for sampling the spent nuclear fuel (SNF) stored in the 105-K Basins (105-K East and 105-K West). This strategy will support decisions concerning the path forward SNF disposition efforts in the following areas:

1. SNF isolation activities such as repackaging/overpacking to a newly constructed staging facility
2. Conditioning processes for fuel stabilization
3. Interim storage options.

This strategy was developed without following the Data Quality Objective (DQO) methodology. It is, however, intended to augment the SNF project disposition DQOs and the Examination Plans/DQOs. The SNF sampling is derived by evaluating the current storage condition of the SNF and the factors that effected SNF corrosion/degradation.

1.2 BACKGROUND

Approximately 103,000 assemblies of N Reactor fuel are stored in the 105-K East and 105-K West Basins. About 1,150 MTU of the fuel are stored in 3,666 open canisters in K East and 958 MTU are stored in sealed, vented canisters in K West. In addition, each basin contains a small amount of Single Pass Reactor (SPR) fuel. Details of the current SNF characteristics and storage condition are given in Appendix A of the "Characterization Plan for Hanford Spent Nuclear Fuel" (Abrefah 1994). The fuel stored in the basins has corroded and degraded to varying degrees, and continues to deteriorate.

DOE/WHO needs to characterize the SNF stored in the 105-K Basins to determine the physical, chemical, and radiological properties of the fuel. The information gathered from the characterization activities will help the DOE/WHO to assess the extent of the corrosion and degradation of the SNF, and to determine those characteristics affecting the various interim storage options being considered by the Hanford Spent Nuclear Fuel Project. Additionally, the information obtained in the characterization activities will support future decisions on final disposition of the SNF.

1.3 SPENT NUCLEAR FUEL CORROSION/DEGRADATION

Uranium fuel, with breached cladding, corrodes when it is contacted by water. The corrosion process proceeds by the following reaction:

Part of the hydrogen generated by the reaction (1) can further react with uranium metal to form uranium hydride by the reaction:

The uranium hydride formed from Equation (2) is very reactive with either water or oxygen. Under the current storage condition most of it can convert to uranium oxide by the reaction:

The zircaloy cladding will also react with water by the process:

Reaction (4), however, proceeds slowly at ambient temperature and is not expected to contribute significantly to the SNF corrosion in the 105-K Basins. In the presence of oxygen, uranium dioxide and uranium hydride can undergo secondary reactions such as:

The above corrosion reactions of the breached fuel element will result in the formation of uranium hydride, oxides of uranium, and hydrogen gas from metallic uranium reaction with water. Small fractions of zirconia and zirconium hydride might be present from the zircaloy reaction with water [Equation (4)]. Due to reactions (3), (4), and (5), the amount of UH_3 present is expected to be small, however, the uncertainty associated with UH_3 reaction mechanism in the storage environment and its high reactivity have raised potential safety concerns in post-storage handling and transportation activities. One of the goals of the characterization program is to determine the fuel corrosion rate and identify the products formed.

Uranium hydride and oxides have densities lower than metallic uranium. These products will increase the fuel volume, which causes additional cracking of the cladding. This process can increase the corrosion rate of the fuel since additional metallic uranium can be exposed to the environment.

Some of the literature suggests that water corrosion of metallic uranium in oxygen-depleted atmospheres is more severe than in aerated water (Wilkinson 1962) British and French experience indicates that an oxygen-deficient environment similar to that of the K West storage mode could enhance uranium corrosion and increase uranium hydride formation. The formation of uranium hydride by gas phase corrosion of metallic uranium clad in steel was

recently reported by Lawrence Livermore National Laboratory (Wood 1994). This suggests possibly greater corrosion in the closed canisters (K West Basin), which contain deoxygenated water and in some cases with the fuel elements extending above the water level in the sealed canisters compared to the open canisters (K East Basin). Therefore, the examination of the fuel stored in K West Basin is a high priority of the characterization campaign.

2.0 SAMPLE VARIABILITY

2.1 VARIABLES POTENTIALLY AFFECTING CORROSION

A number of factors might have contributed to the degradation of the spent nuclear fuel stored in the basins. Some being considered in selecting fuel samples for characterization activities include:

1. Breached cladding: The fuel inventory contains elements whose cladding was breached during reactor discharge and/or subsequent handling. The cladding failures range from cracks to broken fuel elements. A large number of the failures occurred at the end of fuel elements due to collisions with other elements during discharge and subsequent handling in the discharge chute and basins. The element end cap braze joints of the fuel element might also have failed due to the combined effect of irradiation and subsequent corrosion in the basins. These created a variety of effective surface areas that can affect degradation rates.
2. Storage mode: There are two major storage modes; open (K East Basin) and encapsulated (K West Basin) canisters. The open canisters include both closed and open bottoms (i.e., screen bottoms) in the K East Basin. The uranium corrosion rate is strongly influenced by the storage environment. In the sealed canisters in K West Basin a large number of fuel elements had portions of the fuel extending into the gas space of the canisters. This condition presents a possibility of gas phase oxidation of the K West SNF. In addition, the enclosed atmosphere could deplete oxygen (by reaction with exposed fuel) in the canister water, changing the chemistry of the water and potentially accelerating the corrosion of the fuel.
3. Irradiation history: The irradiation histories of the SNF can be divided into two classifications by the isotopic composition of ^{240}Pu . Most of the fuel has been classified as either 6% ^{240}Pu (i.e., weapon grade material with burnup of 907 and 1,098 MWD/MTU for Mark IV and Mark IA fuels, respectively) or 12% ^{240}Pu (i.e., fuel grade material with burnup of 2,268 and 2,720 for Mark IV and Mark IA fuels, respectively). Irradiation history affects the fuel degradation mechanisms mainly through fuel swelling and embrittlement. Fuel swelling and embrittlement can generate microcracks which are potential reaction sites for uranium oxidation/corrosion.

4. Fuel type and enrichment: There are three main types of spent fuel in the basin; N Reactor Mark IV, Mark IA, and SPR (which constitutes a minor fraction of NSF inventory). The fuel types vary in sizes and degree of ^{235}U enrichment. Most of N Reactor Mark IV elements have the potential to be above the water level in the sealed K West canisters and may have experienced vapor phase corrosion. Most of the N Reactor spent fuel stored in the basins have an enrichment of 0.947% ^{235}U with the exception of the outer tubes of the Mark IA fuel elements which have 1.25% ^{235}U and a small inventory of Mark IV assemblies (outer and inner elements) with 0.71% ^{235}U . The SPR fuel was primarily made in two ^{235}U enrichments; natural (0.71%) and 0.947%. Other enrichments were manufactured which included; 1.25%, 1.7%, and 2.1% ^{235}U . Enrichment affects fuel burnup and thus embrittlement and its corrosion characteristics.
5. Galvanic effects during storage: Two materials were utilized for the SNF canisters; aluminum and stainless steel. The potential galvanic coupling of zirconium and/or uranium with the storage containers may affect the degradation of the fuel. SPR fuel elements with aluminum cladding could present different corrosion characteristics. Aluminum, for example, is reported to be anodic to uranium at the initial stages of water corrosion. The coupled uranium may have a higher corrosion rate than an uncoupled uranium.
6. Basin water quality: Over the years the water quality of the basins has varied. At one point in time chlorine was added to the basin water as a fungicide. This practice was stopped when evidence of pitting corrosion was seen on the aluminum storage canisters. The current practice is to utilize demineralized water. Since the SNF storage spans a range of time, different fuel groups have been exposed to different qualities of basin water.
7. Storage time: The length of time the fuel has been in wet storage directly affects the current condition of the fuel. Fuel with breached cladding will experience more corrosion with increased storage time. Also fuel with long storage histories have experienced considerable handling, which could have mechanically damaged the fuel cladding. This has the potential of increasing the amount of metallic fuel available for corrosion.
8. Fuel assembly configuration: The N Reactor fuel assembly is made of two concentric tubes held together by locking spacers. During discharge and handling the outer elements experienced much more mechanical damage than the inner tubes. It is likely that the outer elements experienced more fuel degradation than the inner elements.

The selection of characterization samples for the SNF inventory must take into account these factors and the total population of fuel stored in the basins. The large number of fuel assemblies (~103,000) in the K East and K West Basins, their different storage modes and histories, makes a statistical sampling process impractical from the standpoint of cost and schedule. The statistical sampling of the fuel is further hindered by the high cost involved in working with radioactive materials. Thus, for the

characterization activity to provide data that supports the decision-making process, a phased approach has been adopted. In this approach, the initial samples will be chosen to assess the general condition and physical properties of the SNF. Once this data has been obtained, it will be evaluated and utilized to refine the sampling plan.

2.2 MAJOR VARIABLES

In order to efficiently characterize the fuel, it will be necessary to reduce the number of variables that have been identified to effect the SNF degradation. The characterization program will evaluate those variables considered to be essential to the condition of the SNF in the basins in a timely manner to support the path forward decisions. Other variables will be contemplated as time and funds permit. The following are the primary variables considered to effect the degradation of the SNF, and are the variables selected for investigation during the initial phase of fuel sampling:

1. *Breached cladding*: Provides the pathway for uranium-water corrosion and degradation.
2. *Storage mode*: There are two major storage modes; open and encapsulated canisters. However, there are several subsets of each of these (i.e., closed bottom, open bottom, stainless steel, and aluminum canisters).
3. *Storage time*: The SNF storage time in the basins ranges between 5 and 23 years.
4. *Irradiation history*: There are two major irradiation histories for the SNF; 6% ^{240}Pu and 12% ^{240}Pu .

3.0 CANISTER SELECTION

3.1 K EAST BASIN

A screening process using the results of video records of the spent fuel inventory in the K East Basin in conjunction with existing databases (e.g., irradiation and storage history; reports, data sheets, video tapes, and memos relevant to fuel discharge activities, subsequent handling and storage treatments; and fuel attributes from the fabrication processes) will be utilized to select canisters. The information generated from these analyses will be used to select fuel elements from the K East Basin. If possible, the selected canisters will be emptied onto an examination table in the basin. This will allow a more detailed inspection of the fuel elements prior to selecting samples for shipment to the hot cells.

3.2 K WEST BASIN

Depending on equipment availability, the sealed canister gas and liquid samples analyses will be performed prior to specifying SNF samples for hot cell examination. Sampling of both the gas and liquid contained in the canisters will provide data relating to the relative degradation of fuel elements in the canisters. The potential uses for gas and liquid samples are:

1. Gas sampling will provide data relating to the amount of hydrogen and gaseous radionuclides (e.g., ^{85}Kr) present in the head space. If no hydrogen is present, then either the canister seal has failed or the fuel has not corroded significantly since it was encapsulated. Additionally, the ratio of nitrogen to hydrogen (taking into account hydrogen generation by radiolysis) could help determine gross fuel corrosion rate.
2. Liquid sampling will help determine the extent of damage in a particular canister by providing estimates of the soluble fission product concentration. Since the canisters are sealed, the soluble fission products released during the corrosion process can be related to the extent of the fuel degradation.

Additionally, a non-destructive examination (NDE) of the SNF in the sealed canisters using techniques such as ultrasound will help determine fuel degradation status. The data, if available, could be used to identify canisters for fuel sample selection. Pilot scale testing of NDE techniques may be performed/required (the 305 Building is currently available for this purpose) to ensure that reliable data can be obtained in the K West Basin.

Current restrictions on the handling of fuel in the K West Basin will not allow the dumping of a fuel canister. This will limit the ability to sample canisters that contain fuel elements with significant swelling.¹ Therefore, the first shipment from the K West Basin will consist of elements that show no visible sign of significant swelling. If evaluations determine that it is necessary to obtain fuel samples from the canisters that contain fuel with significant swelling, an alternative sampling plan will be developed. This could involve the transport of a canister to the K East Basin for extracting the swollen elements.

4.0 PROPOSED SNF SAMPLE SELECTION CRITERIA

The large number of fuel assemblies and the variables represented by the SNF inventory in the 105-K Basins make it unnecessarily costly and programmatically impractical to analyze SNF samples to accurately determine every characteristic of the SNF. The sample selection approach, therefore, is

¹ Significant swelling is defined as swelling that would prevent the removal of the fuel elements from the top of an opened canister.

to minimize the number of characterization samples that will support the path forward decisions by providing bounding conditions of the SNF for: (a) the repackaging, (b) transportation, (c) treatment/conditioning processes, and (d) storage options. The proposed sampling strategy will provide valuable insight concerning the physical and chemical conditions of the fuel in the K West Basin since encapsulation circa 1982 and the visually degraded fuel in K East Basin. In selecting the fuel for characterization, general information on metallic uranium oxidation/corrosion available in the literature and the historical database on the N Reactor SNF were reviewed. In addition the results of visual examinations in the basins will be used. Based on these, the following types of fuel have been identified for the initial phase of characterization:

1. *A breached² Mark IV type fuel element with length codes S or E from K West Basin:* Sampling an element that extends above the water level in the closed canisters and might have undergone vapor phase corrosion will help determine whether significant degradation of the SNF has occurred in the closed canisters and whether any stabilized hydride is formed during the gas-phase corrosion.
2. *A K East Basin outer fuel element with breached cladding:* Since there probably are more outer fuel elements with breached cladding than inner elements, an outer element better represents fuel with damaged cladding. The outer elements, having a greater quantity of uranium than the inner elements, could provide a more bounding example of fuel degradation.
3. *A visually intact K West fuel element:* The selection of a fuel element with cladding intact at the time of encapsulation may be made based on the existing database for the circa 1982 encapsulation. The intact element could provide a bounding example of the fuel degradation in the closed canister environment of the K West Basin if it shows any form of fuel degradation due to storage in the oxygen-depleted water.
4. *Fuel grade (FG) and weapon grade (WG) materials from both basins:* The SNF inventory contains more FG (12% ²⁴⁰Pu) material than WG (6% ²⁴⁰Pu) material, and the FG has been in storage longer than the WG. The FG material with higher burnup is expected to degrade faster than the WG material due to defects and fission-gas accumulation in the fuel matrix. Comparisons of WG and FG fuel will help in determining whether this factor will be significant in the next phase of fuel sampling.

² Breached cladding refers to failures such as small pin hole, cracks and completely broken fuel element.

Based on the results of the basin visual examinations and the characterization results to be obtained for these samples, some or all of the following types of SNF will be selected for characterization in the subsequent fuel sampling phases:

5. *Breached fuel elements with the shortest and longest storage time:* The corrosion rate of the spent fuel in the basin might be dependent on the storage history. The variation of the storage history in the two basins can be analyzed by comparing fuels with the long and short storage times.
6. *A visually intact K East fuel element:* This sample will show whether an intact fuel element is free of the corrosion/degradation processes. Intact, uncorroded fuel may also be used to study the corrosion kinetics of irradiated uranium. Additional analysis, such as, radiochemistry and mechanical integrity testing could be performed with this sample.
7. *The single pass reactor fuel:* The SPR fuel with the longest storage history has aluminum cladding, which can influence both the corrosion rate and corrosion products. The zircaloy cladding of the N Reactor fuel, unlike aluminum, is a possible sink for hydrogen generated by the uranium corrosion. This might influence the amount of hydride, if any, formed in the two kinds of fuel, and provide a basis for comparison with uranium metal fuel hydriding found in the literature.
8. *Fuel from a "chip can":* These are broken pieces of fuel with a relatively high surface of exposed metallic fuel. The extent of corrosion will provide information regarding the effect of increased surface area on the corrosion rates. Examination of the "chip can" fuel samples could quantify the time required for N Reactor fuel pieces to corrode completely to an oxide form. Quantification of corrosion rates of fuel pieces will be important information in the event that uranium oxide is chosen as a suitable interim storage form for the SNF.

Sludge from all the selected canisters, when found in quantity sufficient to be analyzed, should also be examined.

These samples will help establish what types of fuel conditions that could represent the worst case conditions for the various interim storage concepts of the SNF, establish a sampling strategy in support of characterization for extended interim storage, and support future analyses for final disposition of the fuel. Additional sampling will be required to resolve all the issues relevant to the path forward decisions concerning repackaging activities for the staging facility, conditioning and process selection for the long term interim storage, and final disposition. The characterization data from these fuel samples will provide inputs into the disposition decision DQO process for directing the examination plan in selecting next batch of fuel samples.

5.0 SUMMARY

Due to the number of fuel elements in the 105-K Basins and many other factors effecting their current condition, a conventional statistical approach will not be used to select samples for the characterization activities. Statistical methods, however, will be used to analyze the video taped results of the SNF in the K East Basin. On the basis of these analyses, canisters from which fuel samples will be chosen for characterization activities, will be selected. Statistical tools will also be used in the testing of specimens (i.e., cut fuel pieces) in the hot cell. It is therefore necessary to develop a fuel sampling strategy that has the potential of reducing sample variability due to the effects of storage environment and at the same time provide the necessary information needed to support the path forward decisions.

To accomplish the fuel sampling objectives, a phase approach is proposed for selecting SNF samples from the K East and K West Basins. The first phase will identify the canisters from which fuel samples can be selected. The second phase will select the specific fuel element samples for characterization activities.

Phase I: Canister selection

In the K East Basin, the visual inspection data will be used for determining which canisters will be selected for sampling. The historical database will be used to help identify the type of elements called for by the examination plan.

In the K West Basin, the encapsulation records will be reviewed and a limited in situ program of gas and liquid sampling should be utilized to quantify gross canister conditions.

Phase II: Fuel sampling

Once the canister selection effort has been completed, the following types of fuel samples from the two basins (unless indicated otherwise) are proposed for shipment to the 327 Building for characterization:

- *A breached Mark IV type fuel with length codes S or E from K West Basin*
- *A K East Basin outer fuel element with breached cladding*
- *Visually intact fuel element*
- *Fuel grade (FG) and a weapon grade (WG) material (breached and intact elements)*
- *Breached fuel elements with the shortest and longest storage time*
- *Fuel from the "chip can"*
- *Single pass reactor fuel.*

The fuel samples should be selected in the order of priority listed, unless certain programmatic redirection and/or the path forward decision DQO suggest otherwise. These samples will provide data for determining SNF worse case conditions and establish bounding characteristics of fuel in the two basins.

The associated sludge in the canisters from which fuel elements have been selected for characterization must be sampled for testing. Because the nature of sludge contained in the canisters has direct relation with the SNF degradation.

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