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Multi-Canister Overpack Pressurization Monitoring and Control Methodology for the Spent Nuclear Fuel Project

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Abstract: A control methodology is developed and monitoring alternatives evaluated for controlling pressurization in a Multi-Canister Overpack for the Hanford Spent Nuclear Fuel Project. Monitoring alternative evaluations include concept description, identification of uncertainties, and identification of experimental work required for implementation. A monitoring alternative is recommended and implementation requirements, risks and start up testing associated with the recommendation are discussed.

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MULTI-CANISTER OVERPACK PRESSURIZATION MONITORING AND CONTROL METHODOLOGY FOR THE SPENT NUCLEAR FUEL PROJECT

EXECUTIVE SUMMARY

Maintaining the Multi-Canister Overpack (MCO) as the primary fuel containment barrier by avoiding over pressurization is a fundamental safety argument for the SNFP. Therefore, a control scheme for precluding MCO pressurization is required to ensure operations are within a defined safety envelope. The current project baseline identifies two points where the MCO is in an unvented configuration and at risk to over pressurization:

- during transport from Cold Vacuum Drying in the 100 Area to the Canister Storage Building in the 200 Area, and
- during interim storage in the Canister Storage Building after processing by the Hot Conditioning System.

Residual water represents the primary source of pressurization in the MCO when in an unvented configuration. During transportation, the residual water of concern is hypothesized to be in the form of free water pockets constrained by diffusion paths and hydration water associated with residual corrosion product. During interim storage, the residual water of concern is hypothesized to be primarily chemisorbed (surface absorbed) water and hydration water associated with a smaller fraction of the residual corrosion product. The chemisorbed water is present during transportation, but is not projected to be available for reaction with fuel at the temperatures experienced by the MCO during transport.

The selected control methodology can impact the risk of achieving program goals (cost and schedule) during operation. Bounding arguments which do not rely on measurement systems for pressurization control represent a control method containing minimal programmatic risk, as safe operation is an inherent operating characteristic. Attempts to create bounding arguments for precluding MCO over pressurization based on conservative assumptions, eliminating the need for a control system, were unsuccessful. Therefore, it was concluded that some type of monitoring system which measures an attribute for each MCO produced is required to demonstrate operational control.

The recommended control methodology is based on establishing safety compliance control points immediately prior to establishing a MCO in an unvented configuration. This results in performing a measurement of the selected pressurization attribute at the conclusion of Cold Vacuum Drying and Hot Conditioning System activities. Since residual corrosion product contributes to the residual water of concern at both safety compliance control points, the likelihood a MCO passes the safety measurement for pressurization control is

dependent on fuel cleaning performance. The ability to physically adjust the characteristic of materials loaded in a MCO is limited to the fuel retrieval system. Identification of a MCO which does not pass the safety compliance test will require performance of one of the mitigation responses identified in Section 5.0. Each of these responses can impact program cost or schedule. Therefore, the rigor of monitoring fuel cleaning performance is related to the programmatic risk that can be accepted during operation.

The recommended control methodology does not rely on a quantitative measurement of fuel cleaning performance to demonstrate that each MCO is likely to pass the safety compliance control point for pressurization. Cleaning process validation tests during start up and periodic sampling were assumed to reduce programmatic risks to an acceptable level. While this approach does not compromise the safety of operations, the potential exists that adjustments to fuel processing systems may be required during start up in order to meet schedule and throughput requirements.

Two basic monitoring alternatives were identified for testing the MCO prior to transport which rely on different bases for completing the safety analysis. These alternative bases are

- Limit the MCO water inventory such that over pressurization is precluded independent of the time required for shipment, or
- Limit the rate of reactions within a MCO such that over pressurization is precluded as long as the transfer is completed in a predetermined time period.

Four alternatives were considered for determining the water inventory of an MCO prior to transport. The alternatives consist of

- combining process measurements at two distinct points in the Cold Vacuum Drying cycle with equilibrium data derived from laboratory data to infer the hydrated corrosion product and hydration water inventory,
- direct measurement of the MCO hydrogen inventory using an instrument based on neutron interrogation of the MCO contents, inferring the water inventory from the hydrogen inventory (similar applications in use, but significant development likely to be required to prove the concept is viable for this application).
- a comparison of the measured bulk fuel density with the theoretical density of intact fuel to infer the quantity of corrosion product remaining with an assembly after cleaning, inferring the MCO water inventory from the corrosion product inventory, and

 a comparison of measured weight of fuel loaded in a fuel basket for comparison with the theoretical weight of intact fuel to infer the quantity of corrosion product included in the basket, inferring the MCO water inventory from the corrosion product inventory.

Based on a comparison of identified uncertainties, it is recommended that the MCO pressurization control prior to transport utilize a comparison of measured reaction rate instead of one of the water inventory measurement alternatives. The reaction rate method relies on determining the uranium reaction rate in a MCO at the conclusion of Cold Vacuum Drying, as indicated by a measurement of the hydrogen generation rate and water vapor pressure. The measured uranium reaction rate is compared with a bounding rate determined in the safety analysis of transportation activities that are completed without assuming a specific water inventory limit. Preliminary estimates of implementing this approach indicate that a time constraint of 12 to 24 hrs will be imposed on the overall MCO transfer, in addition to requiring verification that the uranium reaction rate is bounded by a predetermined limit.

Uncertainties exist in the recommended method for monitoring activities at the transport safety compliance control point which still need to be resolved as part of implementation. The major uncertainties requiring resolution are:

- Resolution of the potential for additional reacting surface area to be exposed for reaction with water due to impacts under normal operating and accident conditions.
- Resolution of the potential impact this basis for completing transportation activities may have on safety analyses of the MCO staged in the CSB (No measured MCO water inventory).

Limitation of the MCO water inventory is the ideal monitoring alternative for evaluating the safety of placing an unvented MCO into interim storage. The alternatives identified for measuring the water inventory prior to storage are:

- direct measurement of the MCO hydrogen inventory using neutron interrogation as described above, and
- addition of a known hydrogen isotope spike to the MCO vapor space, inferring the MCO hydrogen inventory based on achieving equilibrium isotopic exchange throughout the MCO materials, and inferring the water inventory from the hydrogen inventory.

Each of these alternatives requires additional work to resolve identified uncertainties. The primary uncertainty for the neutron interrogation instrument is whether adequate sensitivity can be provided by the measurement technique in the radiological environment. The primary uncertainty for the isotopic exchange method is whether equilibrium can be established in a reasonable time to be useful in production operations. Therefore, the

recommended approach supporting interim storage is to pursue development of a direct monitoring instrument equivalent to the neutron interrogation system in parallel with more detailed investigation of the isotopic exchange monitoring method.

If development of both alternatives fails to mature into a system which can be implemented within schedule constraints, the project baseline could be modified to utilize periodic monitoring of the MCO water radiolysis rate over an extended time period prior to unvented storage as a backup position. This could be achieved by deferring final sealing of the MCOs after Hot Conditioning until sufficient data is collected on each MCO to indicate the rate of pressurization from radiolysis is low enough to allow sealed storage for the remainder of the interim storage period.

Implementation of the recommended control and monitoring methodology imposes requirements on the Fuel Retrieval, Cold Vacuum Drying, Hot Conditioning System, Cask/Transportation, and Canister Storage Building sub-projects. These requirements are identified in Section 5.0.

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MULTI-CANISTER OVERPACK PRESSURIZATION MONITORING AND CONTROL METHODOLOGY FOR THE SPENT NUCLEAR FUEL PROJECT

1.0 INTRODUCTION

The Spent Nuclear Fuel Project (SNFP) is preparing systems to move fuel currently stored in the Hanford K Basins to dry storage located in the 200 East Area. The selected concept to accomplish fuel relocation uses a Multi-Canister Overpack (MCO) for primary fuel containment during transport, drying, staging, conditioning, and interim storage (WHC 1995a, WHC 1995c). This MCO is being developed to support an internal design pressure of 150 psig (WHC 1996a), which forms the basis for constraints on pressurization sources whenever the MCO is in an unvented configuration. Maintaining the MCO as the primary fuel confinement barrier by avoiding over pressurization is a fundamental safety argument for the SNFP strategy. Therefore, a control scheme required to ensure that systems operate within an established operating envelope which precludes MCO over pressurization.

Control features are particularly important at points in the fuel transfer process where the MCO is in a sealed (unvented) configuration. As shown in Figure 1-1, this configuration occurs during transport of the MCO from the 100 K Area to the 200 E Area and throughout the interim storage period within the Canister Storage Building (CSB). Further, the DOE has requested an evaluation of sealed staging in the CSB. This indicates the primary control points for verifying the MCO is within a safety envelope precluding over pressurization will be required immediately prior to establishing the unvented MCO configuration, or at the Cold Vacuum Drying and Hot Conditioning System process steps.

1.1 PURPOSE

The purpose of this report is to describe the methodology selected for establishing that the MCOs produced by the SNFP is within constraints established as safety boundaries to preclude MCO over pressurization. In addition to describing the methodology, the evaluation will:

- describe the feasibility/risks associated with the selected monitoring method,
- identify requirements imposed on SNFP sub-projects by the selected monitoring method,
- identify experimental data required to support implementation of the selected monitoring method, and
- recommend start-up testing required to implement the selected monitoring method.

Figure 1-1. Baseline MCO Water Inventory and Configuration at Selected Production Steps



* - Baseline not sealed. Sealed under consideration

1.2 BACKGROUND

The following sources of MCO pressurization have been identified in WHC, 1996b:

- gaseous products (oxygen and hydrogen) generated by water reaction with uranium compounds and water radiolysis,
- initial backfill gas,
- miscellaneous contributions from trace quantities of organic, etc.
- noble gases released as a result of fuel corrosion, and
- helium produced from alpha decay.

Figure 1-2 provides a summary of the materials which may be considered sources of MCO pressurization. The MCO water inventory represents the primary source of MCO pressurization gases, either from radiolytic decomposition or as a reactant with uranium compounds to produce gaseous reaction products. The following discussion provides a description of each pressurization source.

1.2.1 MCO Water Sources

Water can be retained in the MCO as either "free water" or as water chemically bound to solids stored in the MCO, such as residual corrosion product and oxidized fuel. Water chemically bound to solids has formed a chemical union with a substrate material, is not a free liquid, and exhibits a vapor pressure substantially depressed compared to free water. Chemically bound water may be incorporated as waters of hydration, which are distributed uniformly throughout the crystalline structure, or as surface absorbed (chemisorbed) water. Because of the depressed vapor pressure of chemically bound water, it is more difficult to remove through normal drying processes, and the system temperature must typically be raised to 200 °C, or more, to remove it at a rapid rate. Figure 1-3 presents a qualitative comparison of the water vapor pressure associated with the different water sources. The following description of each potential water source is derived from WHC, 1995b.

1.2.1.1 Current MCO Constraint Estimates

Figure 1-1 indicates that the current baseline strategy for transfer of K Basin fuel to dry interim storage contains two points where the MCO will be in an unvented, or sealed, configuration. These points correspond to:

- Transport of the MCO from the 100 K Area to the 200 E Area, and
- Placement of the MCO in the interim storage configuration within a Canister Storage Building tube for an extended storage period (basis is 40 yr, extendable to 75 yr).



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Since water is projected to be the primary source of MCO pressurization, initial concepts for precluding over pressurization have been based on limiting the MCO water inventory as a basis for control. The MCO water inventory allowed prior to transport from the 100 K Area to the 200 E Area is currently estimated to be limited to less than 1.8 Kg. This constraint is based on the initial draft safety analyses for the transport package (WHC 1996d). The initial draft safety analysis relies on the water inventory constraint to conclude that over pressure of the MCO is precluded when:

- no constraints are imposed on the time a MCO is in the unvented configuration during shipping (maximum equilibrium fuel temperature of 131 °C), and
- a hypothesized transport accident engulfs the transport cask in fire, increasing the temperature of MCO fuel contents to approximately 115 °C.

The MCO water inventory allowed prior to placement in interim storage is defined in the Hot Conditioning System product criteria which currently estimates the water inventory must be limited to less than I.67 Kg prior to allowing transfer of sealed MCOs to interim storage in the CSB (WHC 1996b). The product criteria water constraint was developed by identifying constraints for the pressure contributions from backfill gas, organic, noble gases, and helium releases within the MCO during extended storage and allocating the remaining pressure contribution to water sources.

1.2.2.2 Free Water

Free water is defined as water that is present with the MCO as a liquid, is not chemically bonded to any substrate material, and exhibits the normal vapor pressure of water. Free water can form pools of water within the MCO, and also can cling to any available surface through weak physical bonds (absorbed water).

1.2.2.2.1 Bulk Water

Free water can exist in the MCO as bulk pools within the MCO which can be quantitatively removed during Cold Vacuum Drying. Two configurations of pools have been hypothesized, consisting of the drainable water column within the flooded MCO and water held in cupped areas. The drainable water column consists of water in the flooded MCO which can be removed via pumping through the MCO dip leg that extends to the bottom of the MCO.

The MCO is maintained in a vertical orientation throughout the transfer and processing activities. Since the MCO begins the drying process in a flooded condition and is drained from the bottom, the potential exists for cupped areas of metal surfaces (e.g., fuel elements with missing end caps facing up, dished bottom of MCO, and basket bottom surfaces) to trap pockets of bulk water. These water pockets do not have a resistance to evaporation and will be removed quantitatively at the cold vacuum drying conditions.

1.2.2.2.2 Free Water Constrained by Diffusion Paths

The potential exists for free water to exist within the MCO in a configuration which consists of water pockets that exert the normal vapor pressure of water at a local point immediately adjacent to the condensed phase, which must diffuse into the MCO bulk gas space through a mass transfer resistance path. Examples of this configuration include water trapped in a pocket under cladding and interstitial water within corrosion product. If sufficient water exists in this configuration, the normal vapor pressure of water would be observed within the MCO once equilibrium is established. However, long time periods may be required for the system to achieve equilibrium. This condition could also be difficult to differentiate from a leak in the vacuum system

1.2.2.3 Chemisorbed Water

Chemisorbed water is present as a surface phenomena of solid substrates, such as uranium oxide. In the case of uranium oxide, uranium metal ions are surrounded by four oxygen ions with single bonds connecting each uranium ion to each of the oxygen ions. This configuration allows maximum bonding strength and crystalline density to be achieved. Near the planar surface, this spatial symmetry cannot be maintained and considerable distortion of the crystal lattice occurs, leading to large bond strains. As a result of the bond strain, very little energy is required to separate a uranium to oxygen bond, leaving a positively charged empty uranium orbital extending out past the planar surface and a negatively charged oxide orbital. Either orbital can absorb a polar water molecule and subsequent rearrangement leaves two hydroxide moieties at the surface as in:

 $U^{+} + O^{-} + H_{2}O = U^{+}OH^{-} + O^{-}H^{+}$

These hydroxide moieties are bonded very strongly to the oxide surface.

Removal of the chemisorbed water requires the recreation of the highly strained original crystal lattice surface. These hydrogen containing species are the last to be eliminated by high temperature treatment (600 - 800 °C), and the first to reform upon re-exposure to moisture.

In addition to the hydroxide moieties, water molecules can also be surface absorbed and two types of absorption is noted. The first type is hydrogen bonded to the substrate surface which can be removed at room temperature using a vacuum. The second type is oxygen bonded to the substrate surface and can be removed using temperatures in the 200 to 300 °C range under vacuum, or up to 400 °C without vacuum. Hydroxide and water on uranium oxide surfaces have both been observed experimentally through Diffuse Reflectance Infra-red Fourier Transform Spectroscopy.

The quantity of residual water in a MCO resulting from chemisorbed water is directly proportional to the surface area of contained materials. The primary

mechanism for release of the chemisorbed water over the MCO process cycle will be radiolytic decomposition.

1.2.2.4 Waters of Hydration

Waters of hydration differ from chemisorbed water, in that they are uniformly distributed throughout the crystalline phase. The total water content will be much higher with crystals that contain water of hydration than those with only surface water, since the interior volume of the crystal can be filled with water. Water of hydration are oxygen bonded and are typically removed by heating in the range of 200 to 300 °C under vacuum, or up to 400 °C without vacuum.

For implementation on a production scale, removal of hydration water is a function of the vapor pressure associated with the hydrate, the vapor flow rate through the dehydrating material, the vapor flow rate through an aggregate of packed particles (i.e., corrosion product pockets), and heat transfer into the material to disassociate the hydrate. Practically, for materials normally loaded in an MCO, hydrates associated with thin films will rapidly disassociate. Elevated temperatures will be required to remove bound water from corrosion product pockets.

1.2.2.5 Implication of MCO Water Sources

When initially loaded in the basin loadout pit, the MCO is expected to contain water in each of the three states described above. Free water dominates the MCO inventory when the contents are flooded and during the initial draining and drying phases of Cold Vacuum Drying. After Cold Vacuum Drying, chemically bound water is projected to dominate the MCO water inventory due to the presence of residual corrosion product. Basin sludge may contain a variety of metal oxides from fuel corrosion, canister corrosion, or material from environmental sources (e.g., dust). Laboratory analysis of sludge samples taken to date and data from the literature indicated both waters of hydration (called "hydration water" for the remainder of this report) and chemisorbed water are present in basin sludge. Corrosion product is projected to exhibit similar characteristics since it is a component of basin sludge. The relationship between constraints on the MCO water inventory and water content of corrosion product can be translated into a constraint on the residual corrosion product inventory allowed in each MCO.

Bulk water will be quantitatively removed by the operating procedures (draining), temperatures and pressures of Cold Vacuum Drying. Free water constrained by diffusion paths, hydration water and chemisorbed water can be present in the MCO as water sources where the MCO is in an unvented configuration. The relative importance of the remaining water sources is dependent on the water release conditions involved.

For the transport constraint, the MCO system temperature is not projected to exceed 131 $^{\circ}$ C. At these temperatures, chemisorbed water is not expected to be released from its bound state to react with other constituents in the MCO.

However, the water vapor pressure of the MCO gas space could be increased (becoming available for reaction) from water pockets constrained by diffusion paths, as the temperature increase will increase the diffusivity and concentration gradients for mass transfer. An increase in temperature will also increase the equilibrium water vapor pressure of a hydrated solid. At these elevated temperatures, the water vapor pressure could exceed I atmosphere to react with exposed fuel and liberate hydrogen. Therefore, the water sources of concern for the transport control point are hydration water and water constrained by diffusion paths.

The MCO conditions during interim storage are projected to be limited to less than 200 °C. However, radiolysis effects are expected to be observed over the long storage period. Radiolysis is projected to release both hydration and chemisorbed water from the bound state. Water constrained by diffusion paths will be quantitatively removed by the Hot Conditioning System at 300 °C, as the diffusivity is predicted to increase by a factor of two and the concentration driving force for mass transfer increase by a factor of 200 over conditions in Cold Vacuum Drying. Therefore, the water sources of concern for interim storage are hydration plus chemisorbed water.

1.2.3 Backfill Gas

Backfill gases represent a pressurization source a the proposed process point where the MCO is place in an unvented configuration. The backfill gas is required to determine that the sealed MCO, upon assembly, complies with leak rate criteria. The impact of a backfill gas is to reduce the quantity of gas from other sources which can be accommodated by the MCO without over pressurization.

1.2.4 Organic

The degradation of organic materials via chemical reaction or radiolysis may release gaseous products which contribute to pressurization of the MCO. Potential sources of organic entering the MCO include small pieces of foreign debris and oils from process equipment or MCO fabrication. Organic materials are currently projected to be a negligible contributor to MCO pressurization as compared to water reaction products. This hypothesis will need to be confirmed once the methodology for control of water as a pressurization source is selected.

1.2.5 Other Pressurization Sources

Noble gases are released from the uranium fuel as the fuel corrodes. In addition, helium is released during the alpha decay of some transuranics. These are both minor pressurization sources within the MCO. The noble gas releases are directly proportional to the quantity of fuel reacted while the MCO is in an unvented configuration. Helium releases are dependent on the radionuclide content of fuel and the time a MCO is in the unvented configuration.

1.3 FEASIBILITY OF BOUNDING ASSUMPTIONS FOR PRESSURIZATION CONTROL

A preferred method for controlling operations within safety constraints is based on worst case bounding assumptions which indicate compliance with the safety envelope without performing direct measurements. The feasibility of using bounding arguments for water inventory control during transport and storage is considered below. After consideration, it was concluded that bounding assumptions could not be used as a basis for pressurization control for either MCO transport or interim storage and a measured attribute will be required as part of the control methodology.

1.3.1 Bounding Water Inventory during Transport

Since hydration and chemisorbed water is associated with corrosion product, a simple visual observation of residual corrosion product inventory could be proposed as an adequate control method if the quantity of residual corrosion product corresponding to water inventory constraints is impractical to achieve in a MCO under worst case assumptions. Figure 1-4 presents alternative bounding corrosion product estimates for an MCO based on geometric constraint alternatives which range from filling the MCO with corrosion product to filling the annular voids of fuel assemblies with corrosion product. These types of assumptions yield bounding corrosion product estimates which range from approximately 500 to 4000 Kg. Assuming a maximum water content for corrosion product of 10 Wt% after Cold Vacuum Drying, results in bounding estimates for the water inventory by this approach which range from 50 to 400 Kg water. Therefore, this type of bounding argument produces a MCO water inventory which is one to two orders of magnitude larger than the current constraints in use for transport safety analysis (1.8 Kg) and product criteria (1.67 Kg).

Table 1-1 presents alternative bounding descriptions for the geometry of a MCO corrosion product inventory corresponding to a water inventory approximating current constraints (1.6 Kg). The range of bounding descriptions for residual corrosion product in a MCO indicated in Table 1-1 is considered too narrow to be useful in validating that operations comply with current estimates for water inventory constraints. These water inventory estimates based on bounding arguments indicate an alternative attribute must be developed as the measure that the MCO water inventory is within safety boundaries for the unvented transport configuration.





Table 1-1. Bounding Descriptions of Residual Corrosion Product Allowed in a Multi-Canister Overpack (Basis: 8 liters residual corrosion product in a MCO)

Description	MK IV Fuel at 270 Assemblies/MCO	MK 1A Fuel at 288 Assemblies/MCO
Equivalent number of assemblies as cladding shells filled with corrosion product (no corrosion product on remaining assemblies)	5 E length cladding shells filled with corrosion product per MCO (2% of MCO inventory)	7 M length cladding shells filled with corrosion product per MCO (2% of MCO inventory)
Equivalent number of assemblies with internal voids completely filled with corrosion product (no corrosion product on remaining assemblies)	16 plugged E length assemblies per MCO (6% of MCO inventory)	15 plugged M length assemblies per MCO (5% of MCO inventory)
Equivalent length of corrosion product plugging internal void, averaged across all assemblies in a MCO (no corrosion product on remaining assembly surfaces)	Average internal void plug of 3.9 cm (1.5 in) in each assembly	Average internal void plug of 2.8 cm (1.1 in) in each assembly
Equivalent uniform corrosion product film on all assembly surfaces	0.094 mm (0.004 in) thick corrosion product film on E length assemblies	0.11 mm (0.004 in) thick corrosion product film on M length assemblies

Note: Residual corrosion product at volume limit of 8 liters based on 16 Kg corrosion product limit in WHC, 1996, assuming a corrosion product density of 2 Kg/L and 10 Wt% water at the conclusion of Cold Vacuum Drying. Density of these solids is hypothesized to range from 1 Kg/L (UO₃•2H₂O particle density) to 10 Kg/L (UO₃ theoretical particle density) which would impact the above estimates.

a.

1.3.2 Bounding Water Inventory during Interim Storage

Chemisorbed water potentially becomes a major contributor to the MCO water inventory after removal of free water and most hydration water during the drying and conditioning processes. Quantification of the chemisorbed water can be based on the surface area of solids in the MCO. For comparison, alternative surface areas were calculated to be

- MCO Shell Internal Area = 8 m²
- Intact_Fuel Assembly Surface Area in a MCO \doteq 70 to 85 m²
- I Kg Corrosion Product (particulate) = 100 to 10,000 m², depending on specific surface area of material

Therefore, the surface area is dominated by the particulate inventory.

The monolayer of chemisorbed water associated with a uranium oxide surface is estimated to be 3 mg water/m². Figure 1-5 uses this estimate to calculate the contribution of chemisorbed water to the total MCO water inventory, varying the corrosion product inventory and specific surface area of corrosion product. A bounding argument for chemisorbed water control is also indicated on Figure 1-5, where the total MCO volume is assumed filled with corrosion product. Based on this assumption, 1.25 Kg of water would be predicted to be in the MCO as chemisorbed water if the specific surface area of corrosion product is 0.1 m²/g. The bounding argument would allow 0.42 Kg of the water inventory to be allocated to hydration water and eliminate the need for measuring chemisorbed water after confirming the corrosion product surface area is less than 0.1 m²/g.

Unfortunately, the analysis is sensitive to the particulate specific surface area. Olander, 1982 indicates the surface area of UO_2 pellets sintered at 1450 °C is 0.14 m²/g. It does not appear reasonable to expect the corrosion product surface area to approach that of sintered UO₂ pellets. Therefore, it is unlikely a gross assumption that fills the MCO with corrosion product can be used as a bound for the chemisorbed water content.

Since a bounding corrosion product quantity which occupies less than the entire MCO volume is difficult to identify without some type of measurement, it currently appears unlikely that a bounding argument will be available for control of the chemisorbed water in a MCO for interim storage. Therefore, an alternative attribute must be developed as the measure that the MCO water inventory is within safety boundaries for the unverted storage configuration.





2.0 GENERAL PRESSURIZATION CONTROL METHODOLOGY

2.1 CONTROL METHOD REQUIREMENTS

The following requirements have been identified to define the MCO pressurization control method.

Requirement #1 The control method must be capable of determining if the MCO -attribute (such as water inventory) selected for pressurization control is within the constraints defined for handling a MCO in an unvented configuration.

Basis: The control method must demonstrate that the selected attribute is less than the safety boundary constraint. Measurement of a specific attribute value is not necessarily required.

Requirement #2 The control method must be capable of determining an expected attribute value in a MCO with sufficient accuracy such that the error band on measurements does not exceed the attribute constraint.

Basis: Since the control method involves comparison to a safety boundary, the measured attribute plus uncertainty must be determined to be within the constraint in order to allow handling a MCO in the unvented configuration. The measurement accuracy must be sufficient to minimize indicating failure of the drying process when the actual MCO is within the safety boundary.

Requirement #3 The control method must test each MCO produced for comparison against the attribute constraint.

Basis: Section 1.3 indicates a bounding analysis is not available as a control method for precluding over pressurization. Therefore, an attribute must be measured. Due to fuel condition variability, it is unlikely that a case can be made that the condition of a MCO can be inferred from data collected on other MCOs to satisfy a safety requirement.

Requirement #4 The control method must provide a determination that the MCO complies with constraints identified to preclude over pressurization at a point in the process prior to proceeding with handling the MCO in an unvented configuration.

Basis: Operations must be verified to be within the operating envelope established by safety documentation prior to initiating activities.

2.2 GENERAL MCO PRESSURIZATION CONTROL APPROACH

Figures 2-1 and 2-2 indicate the general approach for MCO pressurization control. Figure 2-1 indicates the approach to ensure that MCO pressurization is controlled to comply with constraints derived from the transportation safety analysis, while Figure 2-2 describes the control approach for compliance with interim storage constraints.

2.2.1 Control Approach Supporting Unvented Transportation

2.2.1.1 Safety Compliance Control Point

The control approach indicated in Figure 2-1 is based on establishing an attribute measurement in Cold Vacuum Drying product as a safety boundary control point. The control approach performs the measurement on each MCO as part of the Cold Vacuum Drying operation prior to sealing for transport. This approach satisfies requirements a and 4 identified in Section 2.1. The first two control method requirements are to be satisfied by the selected measurement technique which is discussed in Section 3.0.

2.2.1.2 Process Control Points

As indicated in Figure 2-1, if a MCO passes the safety compliance test, operations can proceed with sealing the MCO for transport. If a MCO fails the safety compliance test, the MCO enters a set of failure response actions described in Section 5.3.1. The response actions available for a MCO failure of the safety compliance test are generally expected to have a substantial impact on the system throughput. This is the result of the test point location which must occur after MCO is in a configuration that makes it difficult to physically adjust the MCO solid constituents (only location to adjust is return to the basin). Ideally, one or more process control points should be provided at intermediate points in the process sequence where a response to an out of range attribute associated with the solid constituents can be responded to prior to MCO loading.

Due to the relationship between MCO corrosion product inventory and water inventory, the fuel cleaning process represents the primary process step which adjusts the MCO solids constituents in support of MCO pressurization control. Therefore, attributes of the products from fuel cleaning (cleaned fuel assemblies and scrap) are included in the control methodology as a process control point supporting the potential for a MCO to successfully pass the safety compliance control point for transport. Since process control points are used only for indication that Cold Vacuum Drying product will pass the safety compliance measurement, qualitative measures of cleaning performance can be considered. The accuracy of cleaning performance measurements become related to the risk the project is willing to accept that a MCO fails the safety compliance measurement after vacuum drying.



Figure 2-1. MCO Pressurization Control Scheme

WHC-SD-SNF-TL-C25 REVC

Figure 2-2. MCO Pressurization Control Scheme Supporting Unvented Storage



2.2.1.2.1 Surface Corrosion Product Control

Visual observations of the fuel condition after cleaning appears to be one practical indication of fuel cleaning performance available at this time. This monitoring approach should be adequate for an indication only process control point. Ideally, based on Table 1-1, a visual observation is required which indicates the cleaning process leaves less than an average of a 1.5 inch length of corrosion product plugging the void region of each assembly. Observation of this type of attribute for each assembly is projected to be highly uncertain due to the remote viewing environment and likely differences in judging corrosion product quantities from operator to operator. Therefore, the following methodology is proposed for indicating the fuel cleaning system supports production of a MCO that will pass the pressurization safety compliance control points.

The proposed fuel cleaning control method is based on in-basin observation of fuel movements which indicate sludge (including fuel corrosion products) is weakly adhered to the fuel and cladding surfaces. The in-basin observation is supported by laboratory observation of the formation of uranium oxide on a metal surface (PNL, 1995). These laboratory observations indicate oxide coatings increase until reaching a thickness of 40 to 50 microns. At this point, the oxides spall off the metal surface and fragment. Due to this weak adherence, it is projected that once an appropriate agitation rate, time of agitation, and water flow rate through the cleaning system is identified, sludge will be reliably removed from the fuel element surfaces.

The weak adherence behavior of corrosion products can be predicted from the respective densities of uranium metal, UO_2 , UO_3 , and $UO_3 \bullet 2 H_2O$, which are approximately 19, 10, 6, and 6 g/cc, respectively. The Pilling-Bedworth ratio identifies the potential for creation of an adherent layer by comparing the ratio of the oxide volume to the volume of metal it replaces. If this ratio is less than 1, the oxide is porous and discontinuous and will not protect the metal (e.g., magnesium). If the ratio is greater than 2, the oxide rapidly spalls off and also produces poor metal protection (e.g., iron). In general, values between 1 and 2 have the potential for creation of an adherent coating (e.g., zirconium, aluminum).

The Pilling-Bedworth ratio for UO_2 is 1.9 which provides a layer likely to be adherent. However, in the presence of oxygen and water, formation of UO_3 , and its hydrates, can occur. These materials have Pilling-Bedworth ratios ranging from 3 to 4, indicating they are not adherent. This sequential density increase of oxide and hydrate layers will also tend to break the uranium corrosion products into fine particles. Finally, the volume growth needed to create uranium hydrate reduces the likelihood that large quantities of hydrate will collect on a fuel element without either failing the cladding or swelling the fuel.

Due to the weak adherence behavior, it is hypothesized that corrosion product bridging physical gaps in assemblies (e.g., the annular water channel between inner and outer elements and the central water channel of the inner element)

will be more difficult to remove by agitation than corrosion product on surfaces. This hypothesis is based on the concept that the weak surface adhesion can be supplemented with compressive forces generated during volume expansion as corrosion products are formed. Therefore, if no corrosion product is observed bridging the assembly void gaps, the control method can rely on the observed weak adhesion properties of corrosion product to conclude an assembly surface has been adequately cleaned for the purposed of process performance indication.

It is proposed that each assembly processed through the fuel cleaning system be visually checked for corrosion product bridging during the start up phase of operations. The observations during start up are used to confirm the hypothesis on corrosion product bridging versus adhesion and adjust the cleaning system operating parameters until bridged corrosion product is eliminated. Subsequent cleaning operations would then proceed by ensuring the cleaning system is operated properly without a visual verification of performance which individually examines most assemblies.

2.2.1.2.2 Trapped Corrosion product Control

A group of assemblies exist within the basins which has corroded to the extent that the basic geometric assembly configuration no longer exists. The group of fuel assemblies is identified in the currently available visual observation data summarized in WHC, 1996c. These observations are limited to describing the one end of fuel that could be viewed in open canisters within the K East Basin. The assemblies viewed represent 72 % of the total number of assemblies in K East Basin. Table 2-1 summarizes the visual observation results.

Category	Percent of Viewed Assemblies in Category	Category Description
Intact	49.2%	No Indication of Cladding Failure
Breached	8.5%	Clad Failure Indicated, but No Visible Reacted Fuel
Defected	37.9%	Clad Failure with Exposed Reacted Fuel Visible from a Portion of an Element Circumference
Bad	4.4%	Cladding Split (with reacted fuel visible around the entire circumference of an element or fuel has corroded to the extent that voids are visible within a shell of cladding

Table 2-1. K Last Basin Visual Observation Summ

Note: Based on WHC, 1996c. Percent viewed in category adjusted to account for viewing only one end. Observations also indicate approximately 5% of canisters contain fuel with stain deposits or crud buildup in vicinity of assembly spacer clips.

Photographs of the "Bad" fuel in Table 2-1, representing 4.4% of the K East Basin assemblies, indicate cladding deterioration to the point that the assembly water flow channels are highly irregular. Additional void regions within cladding are also apparent. Due to the higher probability of corrosion product bridging these regions, it is recommended that a wide range of assembly conditions, including those observed to be in a "Bad" condition, as defined in WHC, 1996c, be processed during cleaning start up tests to reduce the likelihood of transferring pockets of corrosion product into the MCO.

Observation of recent fuel movements performed in support of characterization sampling indicates some defected and bad fuel can be expected to contain fuel oxidation products in the vicinity of cladding splits which, while readily removed by agitation, is confined by a flow path through the split cladding with variable resistance. The cycle time for removal of this material is likely to vary significantly. This characteristic indicates use of a fixed wash cycle time developed by start up test may need to be overly conservative. Therefore, it is recommended that wash cycle monitoring consider the capability to compare the turbidity of water entering and leaving the wash system. This monitoring capability would become one of the operating parameters calibrated by start up test.

2.2.1.2.3 Final Inspection

A final routine confirmation of cleaning performance is recommended based on a visual check of each loaded fuel basket. This visual check is again based on looking for corrosion product bridging the assembly flow channels. With fuel loaded in the fuel basket, one end is visible. If corrosion product is observed bridging an assembly void region after cleaning in this configuration, assuming the flow channels of this assembly are completely filled with corrosion product represents a worst case residual corrosion product in this configuration and still be expected to pass the safety boundary control test. Since viewing is from one end, the final check of cleaning performance could be accepted if no more than 2.5% of the assemblies indicate corrosion product bridging the water flow channels. This translates to no more than one assembly observed with corrosion product bridging an assembly water flow channel in each fuel basket.

2.2.2 Control Approach Supporting Unvented Storage

The control approach indicated in Figure 2-2 is based on establishing an attribute measurement in Hot Conditioning System product as a safety boundary control point. The control approach performs the measurement on each MCO to support the conclusion that Hot Conditioning System is within constraints for unvented storage. Measurements from each MCO satisfies the third and fourth requirements identified in Section 2.1. The first two control method requirements are to be satisfied by the selected measurement technique, which is discussed in Section 4.0.

2.3 ALTERNATIVE PRESSURIZATION CONTROL METHODS

The control methodology and monitoring scheme discussed above supports the current project baseline concept (WHC 1995c). Modification of this baseline would provide the opportunity to consider alternatives to a control scheme for precluding MCO over pressurization during operation of the SNFP systems. One approach to remove the need for safety boundary control points associated with MCO over pressurization is to maintain the MCO in a vented configuration throughout the project processes. This requires revising the transportation system design to allow transport of a vented MCO from the 100 Areas to the 200 Areas and planning to maintain the MCO in a vented configuration throughout the interim storage period, or until gas production abates.

3.0 ALTERNATIVES FOR MONITORING TRANSPORT SAFETY COMPLIANCE CONTROL

Two alternative approaches for maintaining MCO pressurization control during unvented transport are available, depending on the basis selected for safety analysis. The first approach relies on constraining the MCO water inventory included with fuel during transport. The water constraint limits the pressurization sources such that over pressurizing the MCO is not possible, independent of the time used for completing a transfer in the unvented configuration. The second approach for pressurization control is to define a bound for the rate of MCO pressurization combined with a constraint on the time a MCO is allowed to be in the unvented configuration. Monitoring alternatives supporting each of these control approaches are described and evaluated below.

3.1 MCO WATER INVENTORY LIMITATION

Four alternative methods for measuring the MCO water inventory have been postulated to date. These alternatives are summarized as follows:

- The equilibrium data method relies on ensuring the MCO contents have achieved equilibrium at selected conditions, measures the water evolved between condition changes and combines these data collected during processing with equilibrium data from the laboratory to arrive at a measurement of the hydrated MCO water inventory.
- The neutron interrogation method is based on a proposal by SAIC for adapting a moisture sensor using neutron interrogation techniques to arrive at a measure of the MCO water inventory.
- The bulk fuel density method is based on measuring the bulk density of fuel assemblies for comparison with the theoretical density of intact fuel to infer the quantity of corrosion product remaining with an assembly after cleaning. The MCO water inventory is derived from the corrosion product inventory.
- The fuel weight method is based on measuring the weight of fuel loaded in a fuel basket for comparison with the theoretical weight of intact fuel to infer the quantity of corrosion product included in the basket. The MCO water inventory is derived from the corrosion product inventory.

3.1.1 Equilibrium Data Method

3.1.1.1 Description

This measurement method is based on equilibrium data for hydrated material collected in the laboratory. Using a range of hydrated samples, the water content of hydrated material as a function of temperature and pressure can be determined. With these data in place, two equilibrium conditions can be

selected as operating points during Cold Vacuum Drying. After draining free water, evacuating, and heating, the MCO is brought to equilibrium at the first operating point (e.g., 50 °C and 50 torr). Once this operating point is achieved, the MCO is brought to equilibrium at a second set of conditions (e.g., 75 °C and 5 torr) while measuring the quantity of water removed from the MCO. Combining the equilibrium data derived in the laboratory at the two operating points with the quantity of water remaining in each particular MCO. Since the MCO was brought to equilibrium at the second operating point, the equilibrium data can be combined with the residual hydrate quantity to determine part of the MCO water inventory after Cold Vacuum Drying. This approach is depicted on Figure 3-1.

The residual corrosion product quantity in each MCO must be tracked to apply this method after Hot Conditioning System. At this point, the Hot Conditioning System must verify that equilibrium was achieved in the MCO at a third set of conditions (e.g., $300 \,^{\circ}$ C and 5 torr) and the equilibrium data is again combined with the residual corrosion product estimate derived from Cold Vacuum Drying data to determine the MCO water inventory prior to interim storage.

3.1.1.2 Feasibility Evaluation of Equilibrium Data Approach

Establishing the water inventory by the equilibrium method allows prediction of the residual water associated with hydrates. It provides limited insight into residual surface water associated with non-hydrated materials, such as UO_2 . Since no quantitative relationship exists between the amount of $UO_2 \bullet x H_2O$ and the quantity of UO_2 on any given assembly, this method does not allow projection of total residual water. It is credible that all the residual oxide in a MCO is UO_2 , is not hydrated, has residual surface water associated with it, but has no hydrated water. This scenario would yield a false positive indication for compliance with a water inventory. Some other method must be used to quantify surface water; however, since surface water is primarily liberated by radiolysis at a low rate, this method may suffice for shipping.

The feasibility of the equilibrium data approach was evaluated by considering if:

- the uncertainty in MCO water inventory predicted by the monitoring technique is projected to contain sufficient accuracy in discriminating whether or a MCO complies with the safety boundary constraint using instrumentation of realistic accuracy, and if
- other areas of identified uncertainty in the control method implementation gave the potential to be resolved.







MCO Water Inventory After Conditioning = Y₃S

3.1.1.2.1 MCO Water Inventory Uncertainty Measured by Equilibrium Data

A parametric analysis of the predicted MCO water inventory uncertainty using the equilibrium data monitoring method is presented in Appendix A. The parametric analysis is based on an assumed goal uncertainty in the MCO water inventory measurement of 0.1 Kg water when the measured inventory is predicted to be 1.5 Kg water (i.e., water inventory = 1.5 ± 0.1 Kg). The parametric analysis indicates:

- The MCO water inventory uncertainty is more sensitive to the uncertainty in the hydrate equilibrium points than the measurement of water evolved between equilibrium points during the Cold Vacuum Drying phase of control measurements.
- At the operating points assumed in Appendix A, a water inventory uncertainty of 0.1 Kg water can be obtained if the evolved water measurement at Cold Vacuum Drying has an uncertainty of 0.008 Kg and the equilibrium points are measured with an uncertainty of 0.001 Kg Water/Kg corrosion product. The equilibrium points can be measured within the uncertainty by selecting instrumentation identifying the operating pressure and temperature within 1% of the measured value. These measurement uncertainties approximately balance the uncertainty in the predicted MCO water inventory equally among the measurement components.
- The predicted MCO water inventory uncertainty is reduced by maximizing the difference in hydrate equilibrium water content at the two operating points used for measurements during Cold Vacuum Drying.
- The predicted MCO water inventory uncertainty is reduced by selecting the lowest possible hydrate equilibrium water content as the final operating point of Cold Vacuum Drying.
- Assuming instrumentation measurement uncertainty is based on the percent of value (as opposed to a percent of range), the MCO water inventory uncertainty is reduced as the water measured to be evolved decreases.

Figure 3-2 provides a summary of the uncertainty bands projected for MCO water inventory based on a set of assumed measurements on a MCO in Cold Vacuum Drying.

3.1.1.2.2 Other Areas of Uncertainty

In addition to the calculated uncertainty inherent in the monitoring technique used for controlling the process, other areas of uncertainty have been identified which have viable solutions in order for the control methodology to be considered feasible. The following discussion describes the other uncertainties which have been identified to date.



3.1.1.2.2.1 Actual Corrosion Product Composition Unknown

The corrosion product composition uncertainty has the potential to impact safety compliance control point measurements completed at Cold Vacuum Drying.

The equilibrium data methodology currently assumes no effort will be made to verify that corrosion product included in a MCO is of a known composition during routine operation. The equilibrium characteristics of corrosion product is projected to be dependent on the metal oxide composition. The proposed monitoring method relies on generating equilibrium data from sludge samples which provide an upper bound for the equilibrium water content of corrosion product at the process operating temperatures and pressures. As long as the equilibrium data is considered bounding, the measurement technique would over estimate the actual MCO water inventory.

The solution proposed for resolving this uncertainty is based on selecting sludge samples from basin locations projected to contain increased quantities of metal oxides (e.g., Fe_2O_3 and Al_2O_3) that include hydration water removed at higher temperatures as a basis for constructing equilibrium curves used by the monitoring technique. The selection of sludge samples is described in Section 3.1.1.3.2. The hypothesis that equilibrium data from the selected samples bound corrosion product in the MCO could be tested as part of start up activities.

3.1.1.2.2.2 Verification Equilibrium Achieved by Operating Systems

There is the potential for uncertainty in identifying that a true equilibrium has been achieved throughout the MCO. This uncertainty has the potential to impact safety compliance control point measurement at Cold Vacuum Drying.

The equilibrium data monitoring method relies on the system achieving equilibrium conditions within the MCO during operation for comparison with experimental equilibrium data. Any system held at a constant set of conditions for a long period of time will theoretically achieve equilibrium. A system would typically be considered to approach equilibrium when no significant change in the closed system pressure is observed when held at a constant temperature. Corrosion product deposits in occlusions under cladding defects have been observed in basin fuel. This observation indicates the potential exists for tortuous water diffusion paths to result in slow MCO system responses to temperature and pressure changes. If these changes are very slow, it may be difficult to determine when the MCO system is effectively at equilibrium.

The proposed method for resolving this uncertainty is based on performing a small scale test on a single fuel element, or portion of an element, which duplicates the planned operating conditions of Cold Vacuum Drying. The test should include recording the time taken to observe what was assumed to be equilibrium conditions at the operating points and collect data equivalent to that planned in the control methodology. Upon completion of the test phase, the test element should be held at the highest practical temperature and
lowest practical pressure until no weight loss is observed. If the water inventory identified by the final test phase is bounded by that determined from equilibrium calculations, then the test times would be used as the definition of time required to achieve equilibrium during plant operation.

3.1.1.2.2.3 Non-Uniform Temperature Profile in MCOs

The potential exists for temperature instrumentation to yield an inaccurate effective temperature indication for definition of the equilibrium conditions due to non-uniform temperature profiles in MCOs. This uncertainty has the potential to impact safety compliance control point measurement in Cold Vacuum Drying.

The equilibrium data monitoring method relies on measuring the equilibrium conditions within a MCO for application of equilibrium data. While measuring the effective pressure within a MCO is considered straight forward, identifying the effective system temperature could be affected by non-uniform temperature profiles resulting from varying locations of the fuel heat load. The uncertainty in effective temperature measurement is dependent on the location of temperature sensors in the operating system and may be as high as 30 °C radially.

The proposed method of resolving this uncertainty is to rely on thermal models to identify a bounding location for temperature sensors. This assumes that a location (or multiple locations) can be identified such that the effective MCO system temperature for use in the equilibrium monitoring method is demonstrated to be greater than or equal to the temperature indicated by the measurement instrumentation. Based on this assumption, the monitoring method would over estimate the MCO water inventory compared to the safety boundary control constraint.

3.1.1.2.3 Equilibrium Data Method Feasibility Assessment

The calculated uncertainty for the selected control methodology and equilibrium data measurement method is estimated to provide sufficient accuracy in the MCO water inventory given measurement of parameters which are judged to be practical to achieve. In addition, a method can be proposed for resolving each of the other uncertainties currently identified for the measurement of residual hydrated water using this control methodology. Therefore, the control scheme is theoretically feasible according to the criteria defined at the beginning of this section.

However, the method for resolution of some of the uncertainties may be determined to be infeasible to pursue, or provide results that impose impractical equipment design or system capacity requirements. Therefore, it is not possible to assess the selected control methodology as being feasible without qualifications at this time.

3.1.1.3 Experimental Data Requirements

The equilibrium data monitoring method for measuring the MCO water inventory relies on building a set of equilibrium data for the water content of corrosion product. These data are equivalent to corrosion product drying curves which are developed at a variety of system pressures. The following discussion describes the current sludge drying data available and data requirements which specifically support implementation of the selected MCO water inventory control methodology.

3.1.1.3.1 Current Data Available

Figures 3-3 and 3-4 summarize the currently available sludge drying data in the format supporting implementation of the equilibrium data monitoring method. The sludge drying data presented originates from Thermal Gravimetric Analyses (TGA) of K East Basin Weasel Pit and Floor Sludge samples. This measurement technique monitors the mass of a sample as it is heated according to a predetermined temperature program. Figures 3-3 and 3-4 were developed assuming the sample mass at the end of the temperature program (approximately 500 °C) represents the mass of dry sludge in the sample.

The data presented in Figures 3-3 and 3-4 are not directly applicable to the construction of corrosion product equilibrium curves for use in the MCO water inventory control methodology. This is primarily due to the data being obtained at a total system pressure of 1 atm. However, the data are valuable for indicating problems which may be encountered in the future and guide identification of future data collection.

The existing data exhibit a wide spread in the sludge water content, depending on the sludge sample analyzed. This type of data spread could be a major problem for implementing the equilibrium data method for MCO water inventory monitoring. Since the measured water inventory is for comparison against a safety boundary, it is likely that the worst case data would need to be adopted as the basis for plant operation, leading to an over conservative estimate of MCO water content and a potentially high MCO reject rate after drying or conditioning.







Figure 3-4. K East Basin Floor Sludge Drying Data



Potential causes for the spread of existing data are summarized as follows:

- Different sample compositions (mixtures of metal oxides and hydrated metal oxides) are indicated by the data and this type of data range must be accommodated by the control method.
- System may not provide equilibrium data at the experiment operating conditions. The temperature program used to generate these data was relatively rapid (increased at 5 °C/min) and the system may not approach equilibrium at the recorded temperatures.
- While the total system pressure was constant at 1 atm, the partial pressure of water vapor over the sludge sample is unknown.
 Operation at a constant partial pressure of water represents the ideal method of data collection for establishing equilibrium data.
- Data collected at total system pressure of 1 atm (760 torr). The proposed control points rely on process measurement at pressures down to 5 torr. The potential exists for the spread of equilibrium data to be reduced as the operating pressure is reduced.

3.1.1.3.2 Future Data Requirements

Since the currently available data does not support construction of the equilibrium data for the MCO water inventory control method, additional data must be collected for implementation of the monitoring scheme. These data must contain the added feature that it is obtained with sufficient quality assurance pedigrees such that it will be accepted as part of a control scheme verifying operations are within a safety boundary.

If this monitoring method is selected, the first step in defining future data requirements is identification of the type of sludge, or location sludge samples are taken from, which can be considered either representative or bounding the characteristics of corrosion product expected to be included in MCOs. The alternative types of sludge, based on locations in K East Basin, are from fuel canisters, the weasel pit, the sand filter backwash pit, and the general basin floor. It is hypothesized that canister sludge will be the closest representation of residual corrosion product expected to be included in a MCO. This hypothesis is based on the canister sludge being physically transferred to the fuel cleaning system along with fuel such that it is the last sludge in contact with the fuel prior to MCO basket loading.

Fuel canisters in K East Basin are constructed of either aluminum or stainless steel. The alternate construction materials could be inferred to expect some canister sludge has the potential to contain aluminum oxides which contain hydration water that is evolved at relatively high temperatures. Therefore, selection of canister sludge from aluminum canisters could be expected to bound the equilibrium water content characteristic of canister sludge.

In summary, it is proposed that the sludge equilibrium data be developed based on canister sludge samples obtained from aluminum canisters in K East Basin. The selection of these samples as the basis for equilibrium data collection is based on the hypothesis that canister sludge will be representative of corrosion product included in the MCO. A test of this hypothesis would be required as part of a start-up test program.

3.1.1.3.2.1 Near Term Data Acquisition Requirements

With selection of the sludge sample type, equilibrium data can be collected specific to building the equilibrium curves supporting MCO water inventory control. The data collection method continues to be based on Thermal Gravimetric Analysis of sludge samples across a range of temperatures. The data should be collected at a constant partial pressure of water in the system.

The proposed development procedure is based on starting with a single homogeneous sample which is divided into eight sub-samples. The TGA analysis is obtained on duplicate sub-samples holding the partial pressures of water in the system constant at 50, 10, 5, and 1 torr. The temperature program should consist of temperature ramps to a series of hold points to ensure sample masses are obtained after equilibrium conditions are achieved. The temperature hold points proposed are 50, 75, 100, 125, 150, 175, 200, 250, 300, 400, and 1000 °C. The sample should be held at each temperature until the sample mass is constant. At the conclusion of each run, the sample should be held at 1000 °C at the lowest achievable pressure until no change in mass is observed. These final conditions are intended to provide the dry sludge mass which is used as a basis for reducing the experimental data into an equilibrium curve. The final extreme conditions are also projected to quantify chemisorbed water removal from the sludge and indicate the significance of this water source in the MCO.

Development of these data using four sub-samples at each pressure represents the near term data acquisition requirements supporting the MCO water inventory control method. With these data, a firmer assessment of the uncertainties projected in the control method can be developed, acceptance of the equilibrium data monitoring method for safety boundary control can be confirmed, and adjustments to the projected monitoring equipment uncertainties can be identified.

3.1.1.3.2.2 Data Required Prior to Readiness Review

Prior to initiating the readiness review, a complete basis for equilibrium data used to support plant operation is required. As a minimum, this is expected to require completion of analysis of the remaining sub-samples to provide a duplicate set of results. In addition, data from a second sample divided into sub-samples may be required prior to the readiness review to provide sufficient confidence that the basis for safety boundary control has been established.

3.1.1.3.2.3 Alternate Data Acquisition Method

The data acquisition described above requires a significant expenditure of resources. The key objective of the data acquisition is development of equilibrium data supporting the control method which is sufficient to support a conclusion that operations are being performed within safety boundaries. An alternative for developing the required equilibrium data is potentially available using thermodynamic data for the major individual components identified in corrosion product. For this approach, multiple sludge samples would be analyzed to determine a representative composition which bounds material expected to be included in a MCO. The component thermodynamic data from the literature, or non-radioactive tests, can then be used to develop the required equilibrium curves. Two or more TGA tests could then be used to confirm that the accuracy of this alternative approach is adequate for use in the control method.

3.1.2 Neutron Interrogation Method

3.1.2.1 Description

SAIC has proposed development of a moisture detection instrument which could potentially replace the equilibrium data method described above with a direct measurement instrument located in Cold Vacuum Drying. Sensor concepts, which may be adaptable, are reported to have been developed for the U. S. Navy (detecting moisture intrusion in antennas) and U. S. Air Force (detecting aluminum corrosion as $Al_2O_3 \bullet X H_2O$). The approach proposed is based on interrogating the MCO with neutrons from a source, such as 252 Cf, and measuring thermal neutrons backscattered from hydrogen. Comparison of the backscattered neutrons to the fast neutron source determines a measure of the hydrogen contained by a MCO. Additional development is required to determine if this approach could achieve the sensitivity required to be applicable for MCO water inventory monitoring.

3.1.2.2 Feasibility Evaluation of Neutron Interrogation Approach

Issues remaining to be addressed by the feasibility analysis include:

- An assessment of the ability to either discriminate between hydrogen tied up with UH₃ and with H₂O, or a confirmation that the residual UH₃ inventory would not be so high to yield false negative for a majority of the MCOs (inaccurate high water inventory indications).
- An assessment of self shielding effects. Since the technique relies on energy loss of neutrons to detect hydrogen and thermal neutrons have a short mean free absorption path in 1% ²³⁵U, the efficiency of the method to sample more than the surface remains to be established.

 Background neutrons from inherent radioactivity of the fuel and from subcritical multiplication must be considered.

3.1.2.3 Experimental Data Requirements

Experimental data requirements can not currently be predicted with confidence for the neutron interrogation method. It is projected that an instrument development program would be required for this method, as opposed to the development of chemical property data required for implementation of the equilibrium data method. This type of program does have the potential for completing most development in a non-radioactive environment which could result in a lower cost, faster paced development program as compared to equilibrium data development. However, it is likely that some type of final proof testing using radioactive materials would be required at a point in a instrument development program for this alternative.

3.1.3 Bulk Fuel Density Method

Comparison of the measured bulk fuel density to the theoretical density of intact fuel provides a potential alternative for quantifying the amount of corrosion product loaded into a MCO at the basins. The measurement method relies on the significant difference in density between uranium metal (19 g/cm³) and metal oxide corrosion products (7 4 - 5 g/cm³) to estimate residual corrosion product inventories. Appendix C provides a description of applying this method on an assembly by assembly basis. The measured density of an assembly is derived by measuring the weight of an assembly under water and in gas. The apparent mass difference observed by weighing in different fluids can be used to estimate the measured assembly density.

The departure of measured density from the theoretical assembly density is assumed to indicate the presence of corrosion product on, or within, the assembly and this density difference is used to derive an estimate of the corrosion product quantity. The MCO water inventory is finally estimated from the corrosion product inventory by either a bounding estimate for the water content of corrosion product, or from equilibrium data equivalent to that described for the equilibrium data method in Section 3.1.3.

Appendix C also provides an analysis of the instrument sensitivity required to provide adequate measurement accuracy to allow implementation of this method. The analysis concludes that the sensitivity of instruments for weighing an assembly would need to be better than 0.005% differential mass in order to quantify the residual corrosion product with sufficient accuracy to confirm water inventories are within the currently identified constraints. Instruments with this accuracy are not expected to be available for production applications.

In addition to the instrument sensitivity requirements, the method considered contains the following unattractive features:

- The measurement, as described, is completed on each individual assembly
- Equipment required to measure an assembly in air requires installation of bell jar assembly in the bottom of the basin which can be purged of water and reflooded. This equipment will be need to include systems to handle contaminated gases from reflooding.
- Translation of the measured corrosion product quantity to a MCO water inventory continues to rely on development of a bounding quantity for the water content of corrosion product from drying data.

Therefore, the fuel density method does not currently appear to be an attractive alternative for measuring the MCO water inventory.

3.1.4 Fuel Weight Method

The fuel weight method provides another alternative for estimating the quantity of corrosion product in a fuel basket containing apparently intact fuel assemblies of known length. The method relies on comparing the measured weight of fuel in a basket to the basket weight expected if the assemblies were intact with no corrosion, estimating an upper bound for the corrosion product contained within assemblies loaded in the basket. The MCO water inventory is estimated from the corrosion product quantity included in each fuel basket. Appendix D provides a description of the development of this method and an assessment of the associated uncertainty.

The method is applicable if

- All fuel assemblies in the basket are clean and free of surface corrosion product,
- All fuel assemblies in the basket are not swollen by corrosion under the cladding,
- The length and model of each assembly loaded in the basket are known, and
- Accurate tare and loaded weights for each basket is obtained.

Since this method is not applicable to all fuel baskets and relies on conversion of the corrosion product estimate to the MCO water inventory using drying data, it does not currently appear to be an attractive alternative for measuring the MCO water inventory.

3.2 MCO REACTION RATE LIMITATION

3.2.1 Description

The current draft Safety Analysis Report for Packaging (SARP) supporting cask transportation selected an analysis approach assuming the total MCO water inventory is limited such that MCO over pressurization is precluded. This is a conservative approach which places no constraint on the time a MCO is allowed to be in the unvented configuration during transport, but relies on limiting the inventory of reactants (i.e., water) to preclude over pressurization. An alternative analysis approach could be considered which completes the safety analysis assuming an unlimited reactant inventory in a MCO and relies on reaction rates that are slow enough to preclude MCO over pressurization if the time in an unvented configuration is limited. The following discussion presents a description of the assumed SARP analysis basis, control methodology, and uncertainties in the control approach which results from this alternative to MCO water inventory control.

3.2.1.1 Modified Safety Analysis

In order to depart from a MCO water inventory control scheme, the safety analysis for transportation activities must be revised to analyze MCO performance for the bounds of normal operations and accidents assuming an unlimited inventory of reactants. Since the reactants are assumed to be unlimited during the analysis, the heat of reaction can become a significant contributor to the MCO heat load during the analysis of thermal transjents.

A safety analysis based on unlimited reactants is projected to determine reaction rates which can be accommodated in a MCO for a limited time period during transportation. It is projected that the normal transportation cases would establish a relationship between the maximum time for the MCO to be in the unvented transportation configuration and uranium reaction rates. A plot of the relationship can be developed by describing the reaction rate used for calculating transport times by the reaction rate at a standard set of conditions.

Allowable uranium reaction rates would expected to be constrained by an upper bound determined by analysis of accident scenarios, which supply a heat source in addition to radioactive decay, heat of corrosion reactions, and environmental heating assumed for normal shipping activities. Allowable shipping times would be expected to be constrained by a lower bound time for maintaining the MCO in an unvented configuration (including time to remove a MCO from a Cold Vacuum Drying Station, transport to the 200 Area, and receive at the Canister Storage Building, plus contingency to recover from accidents). The safety analysis would have then determined a safety envelope which could be used for selection of an operating boundary.

Figure 3-5 presents a preliminary transient analysis for the MCO/Cask with unlimited water inventory. The cask is assumed cooled to 25 $^{\circ}$ C at the conclusion of cold vacuum drying and the transient peak fuel temperature



Figure 3-5. Preliminary Transient Analysis of MCO Transport with Unlimited Water Inventory

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calculated for three different assumed uranium reactions rates. The cask is then shipped in an environment assuming maximum ambient temperatures of 46 °C with solar radiation on a diurnal cycle. A maximum decay heat of 835 W is assumed to be contained by the MCO and water vapor is assumed to be available to react with uranium metal based on the partial pressure of water at the average fuel temperature within the MCO. The time for the peak fuel temperature to return to 50, 60, and 70 °C are obtained from the transient analysis to construct alternatives for selecting a transport time constraint. The results of the analysis are shown on Figure 3-6, indicating that a transport time constraint in the range of 12 to 24 hrs could be imposed by this basis for the safety envelope.

3.2.1.2 Reaction Rate Monitoring Implementation

The reaction rate limit control approach for unvented transport is based on comparing a measured uranium reaction rate in each MCO with a reaction rate limit derived from the safety analysis evaluation described above. The corrosion reaction of importance during transportation is projected to be

 $U + 2 H_2 O \Rightarrow UO_2 + 2 H_2$ ΔQ = Heat of Reaction, cal/gmole U

A rate model has been identified for this reaction in oxygen free water vapor which takes the following form (WHC 1996e):

For temperatures less than 523 °K,

Reaction Rate
$$(g U/Hr) = 159 A 10^{-\frac{2144}{T}} [P]^{0.5}$$

where A is the reacting surface area in square centimeters, T is the system temperature in $^{\circ}$ K, and P is the system partial pressure of water in kilo-Pascals (kPa).

The reaction stoichiometry and form of the reaction rate model are combined with process measurements obtained from each MCO at the conclusion of cold vacuum drying. Measurement of a steady state hydrogen generation rate is used to establish the uranium reaction rate observed in each MCO. A measurement of the partial pressure of water and temperature associated with the hydrogen generation rate is required to complete the comparison of measured uranium reaction rate with the acceptable bounding reaction rate identified in the safety analysis. The equations used for developing the reaction rate comparison are shown on Figure 3-7.

Figure 3-6. Time Constraints for MCO Transport





Based on this approach, the transport activities would pass the safety compliance control point for transportation shown on Figure 2-1 if

- the measured uranium reaction rate is less than the uranium reaction rate limit,
- the total time a MCO is in an unvented configuration is limited to the time constraint associated with the reaction rate limit, and
- the MCO/Cask is brought to initial conditions assumed in the safety analysis for development of the time constraint a MCO can be in the unvented configuration.

3.2.2 Uncertainties in Modified Transport Control Approach

The following discussion describes the uncertainties identified to date with the approach of measuring uranium reaction rates as a safety boundary control point.

3.2.2.1 Uncertainty in Uranium Reaction Rate Comparison

A complete analysis of the uncertainty for this approach has not been completed. A preliminary calculation based on example values is shown in Appendix B. The example calculation indicates that uncertainty bounds are small enough to determine that a MCO passes the safety boundary control point if measurement uncertainties are approximately 0.1 gmole/hr for the hydrogen generation rate, 2 °K for the temperature, and 0.1 kPa for the partial pressure of water in the MCO. These estimates are provided as an indication of measurement sensitivity required which must be confirmed by a more detailed analysis.

3.2.2.2 Potential for Localized Reaction Propagation

The proposed measurement technique measures a total uranium reaction rate within a MCO without determining the distribution of reacting fuel surfaces. With an unlimited inventory of reactant in the MCO, the potential exists for localized reactions to propagate throughout the MCO from a low initial average temperature if the reactions generating heat are concentrated in a small enough region. The measurement of uranium reaction rates by monitoring the hydrogen generation rate can not discriminate between reactions occurring from surfaces distributed throughout the MCO, or localized in a small region.

The proposed solution for resolving this uncertainty is based on producing a conservative estimate of the relationship between reaction rates and the time a MCO is allowed in the unvented configuration, combined with a hold at temperature time period during cold vacuum drying. The transient temperature estimates developed during safety analysis are based on an assumed reacting surface area concentrated into the fewest possible number of assemblies located in the worst MCO position for dissipation of heat. The transient analysis in Figure 3-5 begins to address this factor by assuming the reacting

surface area is concentrated in 14 assemblies in a fuel basket. However, the implication of scrap baskets have not yet been addressed.

The uncertainty in modeling conservatism can be supplemented by cold vacuum drying observation. This approach is based on holding the MCO in an unvented condition at 75 °C for a time period equivalent to that allowed completion of shipments to the CSB (e.g., 12 to 24 hrs). If minimal reactions are observed after holding at 75 °C, the cask is cooled to a selected temperature (e.g., 25 °C) prior to shipping. The time period allowed for the MCO in an unvented condition during transfer would be based on a thermal analysis which determines the peak fuel temperature has not returned to a temperature which exceeds the 75 °C (e.g., 50, 60, or 70 °C). The rationale for demonstrating that no temperature excursions will be experienced during transportation is based on the thermal mass of the cask precluding a return of the peak fuel temperature to a value and time exceeding that tested at the conclusion of cold vacuum drying. This approach adds a requirement that the MCO be held at 75 °C for a time equivalent to the time a MCO is allowed in an unvented configuration and that the cask be cooled to a specified temperature prior to shipment.

3.2.2.3 Non-Uniform Temperature Profile in MCOs

The potential exists for temperature instrumentation to yield an inaccurate effective temperature indication for relating the measured uranium reaction rate to a reaction rate limit defined by safety analysis. While measuring the composition of gases in the MCO void space (hydrogen and water vapor) is considered straight forward, identifying the effective system temperature could be affected by non-uniform temperature profiles resulting from varying locations of the fuel heat load.

The proposed method of resolving this uncertainty is to rely on thermal models to identify a bounding location for temperature sensors. This assumes that a location (or multiple locations) can be identified such that the effective MCO system temperature defining the measured uranium reaction rate is demonstrated to be greater than or equal to the temperature indicated by the measurement instrumentation, which results in reducing the reaction rate limit used for comparison with the measured reaction rate.

3.2.2.4 Potential for Competing Reactions with Hydrogen

The potential exists for competing reactions to decrease the measured hydrogen generation rate from a MCO which would result in under estimating the measured uranium reaction rate with water. The potential competing reaction identified to date is formation of uranium hydride. The overall formation stoichiometry is as follows:

The proposed method for resolving this uncertainty is to develop a comparison of uranium reaction rates with hydrogen at the cold vacuum drying operating

conditions for comparison with projected hydrogen generations rates from the water reaction based on data in the literature.

3.2.2.5 Potential for Exposing Additional Reacting Surfaces during Transport

The potential exists for exposing additional reacting surface area during transport as a result of normal shipping vibration and/or cask impacts as a result of accident scenarios. An argument for resolution of this uncertainty has not yet been developed.

3.2.2.6 Potential for Localized Reaction Propagation during Staging

Implementation of this alternative has the potential for impacting the analysis of the MCO in staging. At issue is providing an alternative which allows shipping the MCO without confirming that a specific water inventory constraint has been achieved. Without confirmation, it is anticipated that safety analysis of staging in the CSB would need to be based on an essentially unlimited water inventory available in the MCO for reaction. Since the MCO is vented during staging, this is no longer a problem with over pressurization. However, it does create an uncertainty for controlling the potential for localized propagation of reactions due to the inability of the CSB heat rejection systems to accommodate the added reaction heat loads.

The root of this uncertainty is the uncertainty in the distribution of reacting surface area within a MCO. If the reacting surface area distribution was known, the kinetic equations could be used to determine if the measured reaction rate, extrapolated to higher staging temperatures, can be accommodated by the CSB design. Since the distribution is unknown, the measurement of reaction rates in Cold Vacuum Drying at 75 °C can not be extrapolated to the heat generation rate at the maximum CSB fuel temperature of 200 °C with confidence.

One potential method for resolution of this uncertainty is to restrict the maximum MCO fuel temperature during staging to 75 °C by distribution of the staged MCOs throughout the storage array. The feasibility of this approach has not yet been demonstrated by thermal analysis and its impact on the logistics during operation has not been assessed.

3.2.2.7 Reaction Rate Limit Method Feasibility Assessment

The calculated uncertainty for the rate limit method has not yet been evaluated in detail. The example calculation provides a guide for determining if instrumentation is available to achieve the required accuracies. In addition, the potential for competing reactions, implication of exposing additional surface area during transportation, and impact of transport with no quantified water inventory limit on the CSB staging have not yet been adequately evaluated. A judgement on the feasibility can not be made with confidence until evaluation of these uncertainties are complete.

Assuming successful closure of the above items, it appears that the other uncertainties identified have a path to resolution. Therefore, the rate limit method could be considered feasible. There is one remaining area of concern which may impact the desirability of implementing this alternative, and that is the argument developed for precluding localized reaction rates initiating runaway reactions during transportation. Implementation of this argument requires holding the MCO in a vacuum drying station at 75 °C for 12 to 24 hrs, reducing the capacity of a drying station. This may result in an undesirable increase in the number of drying stations required.

3.2.3 Experimental Data Requirements

No experimental data requirements have been identified specific to this monitoring method, assuming the reaction rate equations and thermal models used for safety analysis must be developed independent of the monitoring method selected.

4.0 ALTERNATIVES FOR MONITORING INTERIM STORAGE SAFETY COMPLIANCE CONTROL

The two general approaches for maintaining MCO pressurization control for transport can be applied to control for storage. The primary difference is that the time period of interest associated with the reaction rate limitation approach must consider time periods which extend over many years, rather than hours.

4.1 MCO WATER INVENTORY LIMITATION

4.1.1 Equilibrium Data Method

4.1.1.1 Description

The equilibrium data method for monitoring MCO water inventory at the conclusion of Hot Conditioning System is similar to that described above in Section 3.1.1. Application at the Hot Conditioning System would be limited to establishing a third equilibrium point if the equilibrium data method is selected to support transport safety boundary monitoring.

4.1.1.2 Feasibility of Equilibrium Data Method

The measurement uncertainty analysis described in Section 3.1.1.2.1 and Appendix A is also applicable to implementation at Hot Conditioning System. The analysis was completed assuming the same monitoring method is used at Cold Vacuum Drying and Hot Conditioning System. If the equilibrium data method is applied only at Hot Conditioning System, it is projected the accuracy of instrumentation would need to be improved over the values estimated in Appendix A.

Other areas of uncertainty discussed in Section 3.1.1.2.2.1 and 3.1.1.2.2.3 with respect to corrosion product composition and temperature profiles are also applicable to implementation of the monitoring system at Hot Conditioning System. The uncertainty associated with achieving equilibrium conditions discussed in Section 3.1.1.2.2.2 is not projected to be significant for implementation in Hot Conditioning System since water constrained by diffusion paths is estimated to be removed at the increased temperature conditions.

Inclusion of chemisorbed water as a pressurization source of concern for interim storage results in an additional uncertainty in implementing the equilibrium data method for monitoring at Hot Conditioning System. The equilibrium data monitoring technique does not provide a direct measurement of chemisorbed water in a MCO. This uncertainty does not impact transport considerations, but has the potential to impact safety compliance control point measurements in the Hot Conditioning System.

The equilibrium data monitoring method is limited in measuring chemisorbed water content by the experimental equilibrium data. The operating points used for MCO water inventory control are not expected to remove chemisorbed water.

However, if the equilibrium data from samples is based on a dry sludge mass with chemisorbed water removed, the basis for equilibrium data is demonstrated to bound characteristics of corrosion product, and no mechanism occurs to increase the quantity or surface area of corrosion product in the MCO, then the selected monitoring method would be expected to indicate a bound for MCO water inventory for both hydration and chemisorbed water even though the only hydration water removal is measured.

Unfortunately, the potential exists for the corrosion product inventory to change during staging by uranium slowly reacting with residual water during staging. The potential for this reaction to occur results in an uncertainty in whether the corrosion product inventory has significantly increased between the Cold Vacuum Drying and Hot Conditioning System. In addition, it is unknown at this time if there is a mechanism for residual water vapor or other gaseous constituents to react with the corrosion product itself and alter the corrosion product surface area. Since chemisorbed water is a surface phenomenon, both of these uncertainties have the potential for increasing the chemisorbed water content.

Since a bounding argument which limits the chemisorbed water content is unavailable (see Section 1.3.2), implementation of the equilibrium data method for monitoring MCO water inventory prior to interim storage does not appear feasible.

4.1.1.3 Experimental Data Requirements

If implemented, the experimental data requirements for the equilibrium data method supporting interim storage would be similar to that described in Section 3.1.1.3.

4.1.2 Neutron Interrogation Method

4.1.2.1 Description

The neutron interrogation method implemented to support monitoring interim storage safety boundary control would be similar to that described in Section 3.1.2.1.

4.1.2.2 Feasibility of Neutron Interrogation Method

The primary difference in implementation at Hot Conditioning System is that the opportunity exists for designs to allow access of the instrumentation to the MCO wall, as opposed to performing measurements through both the cask and MCO wall at Cold Vacuum Drying. It has not yet been determined whether this configuration change has a significant impact on measurement accuracy.

4.1.2.3 Experimental Data Requirements

The experimental data requirements discussed in Section 3.1.2.3 are similar to the requirements projected for implementation at Hot Vacuum conditioning in support of monitoring the interim storage safety boundary.

4.1.3 Isotopic Exchange Method

The hydrogen inventory within a MCO can theoretically be measured by introducing a known quantity of hydrogen isotope (either deuterium or tritium) into the MCO vapor space. This approach relies on measuring the quantity of hydrogen in the MCO vapor space, adding a known spike of the hydrogen isotope to the MCO, and waiting a sufficient time period for the hydrogen isotope to equilibrate among all the chemical species containing hydrogen within the MCO. A measurement of the ratio of the hydrogen isotope to the total hydrogen in the gas space can allow derivation of the hydrogen content of solids in the MCO. Assuming all hydrogen in the solids is in the form of water provides an estimate of the MCO water inventory.

Development of the equations used to estimate the water inventory and an example calculation are presented in Appendix E. An assessment of the uncertainties associated with this method has not yet been completed. However, a basic premise for the method to be feasible is that isotopic exchange occurs throughout all hydrogen bearing materials in the MCO in a reasonable period of time. A test program would need to be developed to determine the required equilibrium time period in support of implementation.

4.2 RADIOLYSIS RATE LIMITATION

Development of a MCO reaction rate limit for confirming that over pressurization is precluded during storage is based on the rate of radiolysis producing gaseous products being directly proportional to the radiation field. This produces a linear increase in the MCO gas pressure over time, assuming no significant secondary reactions. A potential method of using this attribute to confirm that a MCO is suitable for unvented storage is to place each MCO in the storage array after hot conditioning and maintain the capability for measuring the pressure build up, or hydrogen generation rate, throughout the first few years of storage (i.e., 5 Yrs). This could be accomplished by deferring final sealing until after the conditioned MCO reaction rate is established or incorporating pressure monitoring instrumentation in each MCO prior to sealing.

Upon establishing that a MCO radiolysis rate is within a bounding rate to allow unvented for the remainder of the storage period (i.e., 35 to 70 Yrs), the MCOs could be cycled through a weld station for final sealing or pressure monitoring could be discontinued.

5.0 RECOMMENDED CONTROL AND MONITORING METHODOLOGY

The following discussion provides a recommended MCO pressurization control and monitoring methodology based on the alternatives considered. The recommendation is based on a consideration of MCO pressurization as an independent control problem at this time. The potential exists that the recommended control methodology for MCO pressurization could be influenced by the results of safety analyses that are not yet complete. These analyses could result in additional safety boundaries to be established that, when integrated with the need to preclude MCO over pressurization, may require revision of the basis for selecting a pressurization control methodology. Examples of the types of constraints that could influence the recommendation include:

- Imposing a constraint on MCO particulate at risk to dispersion, or
- Imposing a constraint on the fuel reactivity that can be safely processed in cold vacuum drying.

5.1 TRANSPORT CONTROL AND MONITORING METHODOLOGY

The uranium reaction rate comparison method is recommended for monitoring pressurization control in support of the transportation safety compliance control point. While this alternative approach contains uncertainties which must be resolved, it appears to be more desirable than the other alternatives.

The equilibrium data method was considered less desirable than the uranium reaction rate comparison due to the need for development of additional experimental data. Once the data is developed from samples, the potential exists for start up test results to indicate that the selected samples used to establish the safety envelope were not bounding. This conclusion leaves open the possibility that start up activities would need to be suspended until more conservative sludge samples were analyzed.

In addition to the schedule uncertainty, the proposed solution for determining that equilibrium conditions are actually achieved during cold vacuum drying may not be considered adequate given the potential for wide variations in fuel condition. Since the equilibrium data method relies on achieving equilibrium conditions at two distinct points during cold vacuum drying, a strong argument must be available as a foundation for the equilibrium data method. It is uncertain that a large enough test program can be completed prior to start up to provide assurance that the presence of free water masked by diffusion paths of indeterminant length has not been masked during the approach to equilibrium conditions during plant operations.

Implementation of the neutron interrogation method for water inventory monitoring during cold vacuum drying would have attractive features if the instrumentation development for this application could be considered complete.

At this time, significant development is required to determine if the sensitivity of the technique is adequate in an environment containing spent fuel and identify an equipment configuration for implementation. Schedule constraints for cold vacuum drying make selection of this alternative for monitoring the transport safety boundary control point unattractive. Since the instrument equipment configuration would be unknown for some time, a space allocation in the cold vacuum drying facility layout would need to be established for implementation. This approach would add uncertainty to a critical path project which was considered undesirable.

The bulk fuel density method described in Appendix C was projected to require instrument sensitivities that would be difficult to obtain from equipment suitable for a production operation. The fuel weight method described in Appendix D was only applicable to a subset of the fuel inventory, leaving the need for identification of a second measurement method to address non-intact fuel. In addition, both of these methods rely on drying data similar to that required for the equilibrium data method to derive a water inventory estimate from the measured MCO corrosion product inventory. Therefore, these alternatives were considered undesirable.

5.2 INTERIM STORAGE CONTROL AND MONITORING METHODOLOGY

The interim storage safety compliance control point is ideally based on a MCO water inventory constraint to support unvented storage. Of the three alternative methods considered to provide this measurement, the equilibrium data method contained an uncertainty that could not be resolved. This uncertainty was the inability to address the inventory of chemisorbed water after Hot Conditioning System. This leaves the neutron interrogation and isotopic exchange methods as alternatives considered with the potential to provide the required monitoring capability.

Implementation of the neutron interrogation capability in the Hot Conditioning System contains the same uncertainty problems indicated above for Cold Vacuum Drying. Due to the lag in start up schedules, constraints for the Hot Conditioning System still contain a small time window to complete sufficient instrument development to determine if the concept is feasible. Therefore, it is recommended that the neutron interrogation instrument development be pursued to determine if the sensitivity is adequate to support Hot Conditioning System product monitoring, identify the equipment configuration footprint requirements, and determine the impact this measurement technique would have on process capacity.

An assessment of the uncertainties associated with the isotopic exchange method is also recommended as a potentially attractive alternative to the neutron interrogation instrument.

If neither the neutron interrogation monitoring or isotopic exchange methods are found to be feasible, modification of the project baseline could be considered to implement the deferral of the MCO storage in the CSB in an unvented configuration until the reaction rate is demonstrated to allow sealed

storage. If the reaction rate monitoring fails to indicate that unvented MCO storage is feasible throughout the storage period, continued periodic venting will be required.

5.3 IMPLEMENTATION RISKS OF RECOMMENDED METHODOLOGY

The discussion of uncertainties in Sections 3.0 and 4.0 indicate that the selected control method was theoretically feasible, but requires resolution of activities to close identified uncertainties before successful implementation can be assured. The uncertainty resolution in itself is an implementation risk for the selected approach. In addition to the uncertainty resolution activities, the potential for false indications by the control methodology was investigated as an indication of implementation risks.

5.3.1 Response Alternatives if MCO Fails Safety Boundary Control Point Test

Figures 5-1 and 5-2 indicate the potential reason for failure and associated response alternatives to failure at the safety boundary control points for transportation (measured during Cold Vacuum Drying) and unvented storage (measured during Hot Conditioning System). The selected control methodology does not currently contain a mechanism for identifying the reason for a MCO failing the safety boundary control point test. Therefore, the only operational options available for response to a test failure would be to follow a response alternative based on an assumed reason. Based on the responses identified in Figures 5-1 and 5-2, it is assumed that the first operational response to test failure in either Cold Vacuum Drying or Hot Conditioning System would be based on assuming equilibrium conditions were not achieved during processing which can be corrected by providing additional time in a processing station. A second response would be consideration of changing the processing station routine operating conditions to increase the temperature and/or decrease the pressure used in processing. Each of these responses would result in an overall decrease in the throughput rate of the process systems while responding to the test failure.

The remaining response alternatives involve returning the MCO to previous process steps to repeat process activities, or moving the MCO forward in the process system past the safety boundary control point. In order to move a MCO forward in the process after failing the safety boundary control point would require implementation of special procedures (e.g., precautions which make the bounding transportation accident incredible) or a major change in implementation plans (e.g., store MCOs vented throughout the interim storage period).

In summary, all responses to a failure indicated at the safety boundary control point results in a major impact on project operations. Due to these impacts, a false indication of failure at the safety boundary control point is highly undesirable. In addition, a false indication of acceptance at the safety boundary control point is unacceptable. Therefore, potential methods of observing both of these false indications were investigated.

Figure 5-1. Alternative Responses to MCO Failing Safety Compliance Test for Unvented Transportation



Figure 5-2. Alternative Responses to MCO Failing Safety Compliance Test for Unvented Storage



5.3.2 Potential for False Indication MCO Fails Control Point Test

Since the control point measurements are associated with safety boundaries, the methodology will always be developed based on conservative assumptions. This results in a planned over estimate of the uranium reaction rate or MCO water inventory which must be balanced against implementing a system that no MCO can pass. Two attributes have been identified which may result in excessive conservatism in the selected transport safety boundary monitoring method.

The first attribute which may result in false failure indications is based on the conservatism incorporated in the safety analysis defining transport time constraints as a function of uranium reaction rate. The monitoring method, as currently proposed, converts measured reaction rates to the same conditions assumed to exist in the MCO by the safety analysis. This conversion could result in some MCOs failing the reaction rate test that could actually be safely transported within the time constraint.

The second attribute which may result in false failure indications is associated with the indicated temperature for an MCO. While not yet confirmed feasible, it is anticipated that a temperature measurement system would be based on the MCO containing heat generating material. At equilibrium, the internal heat generation would provide a temperature profile such that the internal temperature of materials is always greater than the MCO surface temperature. Applying this lower bound temperature to the reaction rate data results in over estimating the MCO reaction rate when corrected to the standard set of conditions used for comparison with bounds derived from the safety analysis.

5.3.3 Potential for False Indication MCO Passes Control Point Test

It is also possible that temperature indications could be obtained which over estimate the effective temperature within a MCO associated with the measured reaction rate. This could result in a reaction rate correction which is nonconservative. Methods for guarding against this type of indication must be developed once the temperature instrument locations have been defined.

5.4 SNFP SUB-PROJECT IMPLEMENTATION REQUIREMENTS

5.4.1 FUEL RETRIEVAL SUB-PROJECT REQUIREMENTS

The following control elements are required to be provided by the Fuel Retrieval Sub-Project for implementation of the recommended MCO pressurization control methodology. These control elements are for indication that adequate fuel cleaning was achieved, decreasing the potential for failing the pressurization monitoring tests at the conclusion of Cold Vacuum Drying and Hot Conditioning System and are considered independent of the selected monitoring system.

5.4.1.1 Determine Fuel Cleaning Parameters During Start Up

Start up plans for fuel cleaning are required to include disassembly/visual examination of cleaned fuel from a variety of starting conditions to establish set points for fuel cleaning parameters that indicate corrosion product bridging assembly water channels has been removed. The fuel cleaning parameters set during start up tests should consider a measure of the sludge leaving the fuel wash system (such as wash water turbidity) to determine the cleaning point. Start up tests should be performed on a wide range of fuel assembly conditions.

5.4.1.2 Provide Capability to Examine Fuel in Loaded Basket

The capability to view a basket of fuel is required to verify that no more than one assembly per fuel basket contains corrosion product which bridges assembly water channels is loaded into a MCO.

5.4.1.3 Prevent Introduction of Sludge into Cleaned Fuel

Reasonable precautions are required to preclude introduction of sludge into fuel baskets during handling. This includes avoiding transfer of canisters over queued baskets or placing baskets on sludge accumulations.

5.4.2 COLD VACUUM DRYING SUB-PROJECT REQUIREMENTS

The following control elements are required to be provided by the Cold Vacuum Drying Sub-Project for implementation of the selected MCO pressurization control methodology. These control elements provide for the comparison of uranium reaction rates at the safety boundary control point supporting unvented MCO transportation.

5.4.2.1 Provide Capability to Measure Hydrogen Generation Rate from a MCO

The capability to measure the hydrogen generations rate from each MCO at the conclusion of Cold Vacuum Drying is required to implement the selected control methodology. The need for redundant measurement of the hydrogen generation rate is unknown at this time. The accuracy of hydrogen generation rates is not considered established, but the uncertainty is on the order of 0.1 gmole hydrogen per hour.

5.4.2.2 Provide Cycle Time for Control Measurements

Time within the Cold Vacuum Drying operating cycle is required to demonstrate that localized reactions will not result in a runaway reaction during the transportation period. Based on the argument resolving this uncertainty in support of the reaction rate limit method, Cold Vacuum Drying time cycles are required to support holding the MCO for 12 to 24 hrs at 75 °C to support this demonstration. In addition, the time a MCO is in the unvented configuration within the Cold Vacuum Drying station in preparation for transport must be

short enough to support completion of the shipment and return of the MCO to a vented configuration at the CSB in support of the shipping constraint.

5.4.2.3 Provide Capability to Measure MCO Conditions

The capability to measure the MCO conditions (temperature and water vapor pressure) during the hydrogen generation rate measurement is required to implement the selected control method. The allowable uncertainty in the measure of these conditions has not been established, but is estimated to be on the order of 2 % for temperature measurement and 0.1 kPa for the water vapor pressure.

5.4.3 Hot Conditioning System Sub-Project Requirements

The following control elements are required to be provided by the Hot Conditioning System Sub-Project for implementation of the selected MCO pressurization control methodology. These control elements provide for the verification that the MCO water inventory is within constraints at the safety boundary control point supporting unvented MCO storage.

5.4.3.1 Provide Cycle Time for Control Measurements

Time within the Hot Conditioning System operating cycle is required to demonstrate that the measured MCO water inventory is less than 1.67 Kg.

5.4.3.2 Provide Capability to Directly Measure MCO Water Inventory

The capability to directly measure the MCO inventory is required to implement the selected control method.

5.4.4 Cask/Transportation Sub-Project Requirements

The cask/transportation sub-project is required to develop a transportation system which allows transport of the MCO from the 100 Area to the 200 Area within the time constraint imposed on the MCO in an unvented configuration associated with reaction rate control.

5.4.5 Canister Storage Building Sub-Project Requirements

The cycle time for receipt of a MCO must be short enough such that when combined with the transport preparation and actual transport, the total time a MCO is in an unvented configuration is less than the selected transport time constraint (12 to 24 hrs).

5.5 SYSTEM START UP TEST RECOMMENDATIONS

The following start up testing recommendations are based on the verification tests identified by the control methodology description in the previous sections of this report.

5.5.1 Develop Fuel Cleaning Parameters

Tests during fuel cleaning start up to set operating cycle times and other parameters are identified in Section 2.2.1.2 to support the control methodology described for fuel cleaning. Completion of these start up tests is directed toward establishing that adequate cleaning of most fuel is achieved to provide adequate confidence that a MCO will pass the safety boundary control points by ensuring the cleaning system was operated within established parameters. The testing then results in most fuel to be loaded in fuel baskets without performing an assembly by assembly inspection.

6.0 REFERENCES

- Olander, 1982 Olander, D.L., D. Sherman and M Balooch, "Retention and Release of Water by Sintered Uranium Dioxide," <u>Journal of</u> <u>Nuclear Materials</u>, 107, pp. 31-43, 1982.
- PNL, 1995 PNL-10778, J. Abrefah, et. al., "K-Basin Spent Nuclear Fuel Characterization Data Report," August 1995.
- WHC, 1995a WHC-SD-SNF-SP-005, (Rev 0), "Hanford Spent Nuclear Fuel Project Integrated Process Strategy for K Basins Spent Nuclear Fuel," July 1995.
- WHC, 1995b WHC-SD-SNF-TI-014, (Rev 0), R. J. Puigh III, et. al., "Technology Status in Support of Refined Technical Baseline for the Spent Nuclear Fuel Project," July 1995.
- WHC, 1995c WHC-SD-SNF-SD-005, (Rev 0), "Spent Nuclear Fuel Project Technical Baseline Description Fiscal Year 1996," November 1995.
- WHC, 1996a WHC-S-0426, (Rev 1), L. H. Goldmann, "Performance Specification for the Spent Nuclear Fuel Multi-Canister Overpack," January 1996.
- WHC, 1996b WHC-SD-SNF-OCD-001, (Rev 0), C. R. Miska, "Spent Nuclear Fuel Conditioning Product Criteria," February 1996.
- WHC, 1996c WHC-SD-SNF-TI-018, (Rev 0), A. L. Pitner, "K East Fuel Cleanliness Evaluation," January 1996.
- WHC, 1996d WHC-SD-TP-SARP-017 (DRAFT), W. S. Edwards, "Preliminary Safety Analysis Report for Packaging (Onsite) Multiple Canister Overpack Cask," April 12, 1996 Draft Version.
- WHC, 1996e WHC-SNF-SD-TI-020, (Rev 0), T. D. Cooper, "Spent Nuclear Fuel Project Recommended Reaction rate Constants for Corrosion of N Reactor Fuel," May 14, 1996.
- Willis, 1995 WHC-SD-SNF-TI-009, (Rev 0), W. L. Willis and A. N. Praga, "105-K Basin Material Design Basis Feed Description for Spent Nuclear Fuel Project Facilities," August 1995.

APPENDIX A

MCO WATER INVENTORY MONITORING FROM EQUILIBRIUM DATA UNCERTAINTY ANALYSIS

UNCERTAINTY EQUATIONS A.1

A.1.1 Cold Vacuum Drying MCO Water Measurement Uncertainty

Based on Figure A-1, the equation determining the residual water content of a MCO after Cold Vacuum Drying is

$$W = A\left(\frac{Y_2}{Y_1 - Y_2}\right)$$

where, W = Residual water content of the MCO, Kg Water A = Water measured to be removed from a MCO between two distinct equilibrium points during Cold

Vacuum Drying, Kg Water Y_1 = Equilibrium water content of corrosion product at the first set of equilibrium conditions during Cold Vacuum Drying, Kg Water/Kg Dry Corrosion product Y_2 = Equilibrium water content of corrosion product at the final Cold Vacuum Drying conditions, Kg Water/Kg Dry Corrosion product at the final Cold Vacuum Drying conditions,

The uncertainty is estimated from

$$\Delta W = \sqrt{\left(\frac{\partial W}{\partial A}\right)^2 \Delta A^2 + \left(\frac{\partial W}{\partial Y_1}\right)^2 \Delta Y_1^2 + \left(\frac{\partial W}{\partial Y_2}\right)^2 \Delta Y_2^2}$$

The partial derivatives of the equation estimating water content are

$$\frac{\partial W}{\partial A} = \left(\frac{Y_2}{Y_1 - Y_2}\right)$$

$$\frac{\partial W}{\partial Y_1} = -\frac{A Y_2}{(Y_1 - Y_2)^2}$$

$$\frac{\partial W}{\partial Y_2} = \frac{A Y_1}{(Y_1 - Y_2)^2}$$

These equations are used to investigate the parametric variation of residual water content uncertainty in Cold Vacuum Drying product.



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A.1.2 Hot Conditioning System MCO Water Measurement Uncertainty

Based on Figure A-1, the equation determining the residual water content of a MCO after Hot Conditioning System is

$$W = A\left(\frac{Y_3}{Y_1 - Y_2}\right)$$

where, W, A, Y₁, and Y₂ are as defined in A.1.1. Y₃ = Equilibrium water content of corrosion product at the final Hot Conditioning System conditions, Kg Water/Kg Dry Corrosion product.

The uncertainty is estimated from

$$\Delta W = \sqrt{\left(\frac{\partial W}{\partial A}\right)^2 \Delta A^2 + \left(\frac{\partial W}{\partial Y_1}\right)^2 \Delta Y_1^2 + \left(\frac{\partial W}{\partial Y_2}\right)^2 \Delta Y_2^2 + \left(\frac{\partial W}{\partial Y_3}\right)^2 \Delta Y_3^2}$$

The partial derivatives of the equation estimating water content are

$$\frac{\partial W}{\partial A} = \left(\frac{Y_3}{Y_1 - Y_2}\right)$$

$$\frac{\partial W}{\partial Y_1} = -\frac{A Y_3}{(Y_1 - Y_2)^2}$$

$$\frac{\partial W}{\partial Y_2} = \frac{A Y_3}{(Y_1 - Y_2)^2}$$

$$\frac{\partial W}{\partial Y_3} = \frac{A}{(Y_1 - Y_2)}$$

These equations are used to investigate the parametric variation of residual water content uncertainty in Hot Conditioning System product.

A.2 PARAMETRIC VARIATION OF UNCERTAINTY

The uncertainty equations in Section A.1.1 were used to investigate the influence of parameter uncertainties on the projected water inventory uncertainty. Parametric variations of the measurement uncertainties and equilibrium point difference were investigated.

A.2.1 Selected Residual Water Uncertainty Goal

A goal uncertainty for the residual water measurement uncertainty was selected for performing the parametric evaluation. This goal was selected as a maximum uncertainty of 0.1 Kg water in the measured residual MCO water content. This goal uncertainty was selected to allow a reasonable approach to a maximum measured residual water content which could be in the range of 1.6 to 1.8 Kg water.

A.2.2 Variation of Measurement Uncertainties

Figure A-2 provides an indication of the change in residual water uncertainty with the uncertainty in the measurement of water evolved between two equilibrium points during cold vacuum drying and the uncertainty in the equilibrium water vapor pressures at the equilibrium points that would be observed based on MCO pressure and temperature measurements. The residual water uncertainty was calculated assuming the measurements indicate a residual water content of 1.5 Kg in a MCO after cold vacuum drying. The equilibrium drying curve was assumed to indicate corrosion product contains 0.1 Kg water/Kg dry corrosion product at the first equilibrium point and 0.05 Kg water/Kg dry corrosion product at the second equilibrium point based on the equilibrium data for weasel pit sludge shown in Figure 4-1 (sample analysis with lowest water content).

Figure A-2 indicates the residual water inventory estimate is reduced as the uncertainty in measurements is reduced. These preliminary estimates indicate the residual water content uncertainty goal of 0.1 Kg water is observed at measurement uncertainties of 2%. Inspection of the contributions to the residual water uncertainty indicates that the uncertainty in the measured water evolved between equilibrium points can be increased to approximately 4% without exceeding the residual water uncertainty goal. This also provides an approximate balance between the contributions of each measurement to the overall uncertainty.

A.2.3 Variation of Equilibrium Point Difference

Figure A-3 provides an indication of the change in residual water uncertainty with the difference in equilibrium operating points assuming measurement uncertainties are maintained constant at 2% for the equilibrium points and 4% for the measurement of water evolved between the two equilibrium points. This variation of residual water uncertainty was developed for an assumed corrosion product water content at the second equilibrium point ranging from 0.01 to 0.1 Kg water/Kg dry corrosion product. The parametric variation was again








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completed assuming the measured residual water content in a MCO is constant at 1.5 Kg after cold vacuum drying. Since the equilibrium point difference impacts the calculation of residual water from the measured water evolved, the assumed quantity of water evolved was varied in order to hold the calculated residual water in a MCO constant for construction of Figure A-3. The assumed quantities of residual water evolved as the equilibrium point difference varies is also indicated.

Figure A-3 indicates the residual water uncertainty is reduced by maximizing the difference in corrosion product equilibrium water content at the two operating points during cold vacuum drying. In addition, the residual water uncertainty is reduced by selecting the lowest possible corrosion product equilibrium water content as the second operating point for cold vacuum drying.

A.2.4 Variation of Measured Water Evolved at Cold Vacuum Drying

Figure A-4 provides an indication of the change in residual water uncertainty as the measured quantity of water evolved between the two equilibrium points at cold vacuum drying increases. Uncertainty bounds for the calculated residual water content in a MCO at the conclusion of cold vacuum drying and hot vacuum conditioning are presented assuming the equilibrium water content of corrosion product is 0.1 Kg water/Kg dry corrosion product at the first cold vacuum drying operating point, 0.05 Kg water/Kg dry corrosion product at the second cold vacuum drying point, 0.02 Kg water/Kg dry corrosion product at the hot vacuum conditioning operating point, and measurement uncertainties selected in Section A.2.2. Uncertainties in the residual water content after hot vacuum conditioning were calculated based on the equations in Section A.1.2.

Figure A-4 indicates that the residual water uncertainty is reduced as the quantity of water measured to be evolved during cold vacuum drying is reduced. In addition, the uncertainty for residual water content after hot vacuum conditioning is smaller than that projected for after cold vacuum drying.

A.3 ESTIMATE OF UNCERTAINTY ALLOWED FOR PROCESS MEASUREMENTS

Based on the parametric variations described above, it appears that process instrumentation which allow measurement of the MCO conditions to allow determination of the equilibrium point with an uncertainty of 2% and water evolved during cold vacuum drying with an uncertainty of 4% of the measured values will support the goal of providing an uncertainty in the MCO residual water content of 0.1 Kg water. The measurement of water evolved is projected to be derived by direct measurements while the equilibrium point is derived from system pressure and temperature measurements. The instrument uncertainty for pressure and temperature measurements can be estimated by assuming the equilibrium data is a general function which varies with temperature and pressure. A linear relationship for this uncertainty is assumed, yielding

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$$\frac{\Delta Y}{Y} = \frac{\Delta P}{P} + \frac{\Delta T}{T}$$

This indicates that an uncertainty of 2% in the equilibrium point can be achieved with an uncertainty of 1% in the measurement of system pressure and temperature.







APPENDIX B

REACTION RATE COMPARISON UNCERTAINTY ANALYSIS

B.1 UNCERTAINTY EQUATIONS

Based on Figure B-1, the MCO will pass the safety boundary control point for transport if $R_{\rm M} \leq R_{\rm LIMIT}$, or

$$f = \frac{R_{M}}{R_{LIMIT}} \le 1$$

where $R_M =$ Measured uranium reaction rate, g U/Hr $R_{LIMIT} =$ Bounding uranium reaction rate adjusted to the conditions (temperature and partial pressure water) that determined R_M , g U/Hr

The uncertainty is estimated from

$$\Delta f = \sqrt{\left(\frac{\partial f}{\partial R_{M}}\right)^{2} \Delta R_{M}^{2} + \left(\frac{\partial f}{\partial R_{LIMIT}}\right)^{2} \Delta R_{LIMIT}^{2}}$$

$$\frac{\partial f}{\partial R_{\rm M}} = \frac{1}{R_{\rm LIMIT}}$$

$$\partial f = -\frac{R_M}{R_{LIMIT}^2}$$

The measured reaction rate derived from hydrogen generation measurements is based on stoichiometry of

$$U + 2 H_{2}0 \Leftrightarrow U0_{2} + 2 H_{2}$$

Therefore,

$$R_{M} = H_{M} \left(\frac{1 \text{ mole } U}{2 \text{ mole } H_{2}}\right) \left(238 \frac{g U}{g \text{mole } U}\right) = 119 H_{M}$$

where H_{M} = gmole H_{2}/Hr measured from a MCO.

The uncertainty in R_{M} is

$$\Delta R_{M} = 119 \Delta H_{M}$$



Based on the reaction rate equation, the reaction rate limit for comparison with the measured reaction rate is derived from the safety analysis bounding reaction rate by

$$R_{LIMIT} = R_{BDD} \left(\frac{P_{M}}{P^{0}}\right)^{0.5} 10^{-2144\left(\frac{1}{T_{M}} - \frac{1}{T^{0}}\right)}$$

Bounding uranium reaction rate selected from safety analysis reported at standard temperature (T¹) and partial pressure water (P⁰), g U/Hr Standard temperature for bounding reaction rate from safety analysis, "K Standard partial pressure water for bounding reaction rate from safety analysis, kPa MCO temperature during measurement of H₂ generation rate, "K MCO partial pressure water during measurement of H₂ generation rate, kPa where R_{BDD} =

T0 = P0 = TM = PM =

 $R_{\text{BDD}},~T^0,$ and P^0 are constants in the uncertainty analysis, therefore, the uncertainty in R_{LIMIT} is

$$\Delta R_{LIMIT} = \sqrt{\left(\frac{\partial R_{LIMIT}}{\partial P_{M}}\right)^{2} \Delta P_{M}^{2} + \left(\frac{\partial R_{LIMIT}}{\partial T_{M}}\right)^{2} \Delta T_{M}^{2}}$$

$$\frac{\partial R_{LIMIT}}{\partial P_{M}} = R_{BDD} \left(\frac{0.5}{\sqrt{P^{0} P_{M}}} \right) 10^{-2144 \left(\frac{1}{T_{M}} - \frac{1}{T^{0}} \right)}$$

$$\frac{\partial R_{LIMIT}}{T_{M}} = R_{BDD} \left(\frac{P_{M}}{P^{0}}\right)^{0.5} \left(\frac{2144 \ln(10)}{T_{M}^{2}}\right) 10^{-2144\left(\frac{1}{T_{M}} - \frac{1}{T^{0}}\right)}$$

B.2 EXAMPLE CALCULATION OF UNCERTAINTY

Assuming the safety analysis bound is determined to be

 $R_{BDD} = 50 \text{ g U/Hr, at}$ $T^{0} = 348 \text{ °K and}$ $P^{0} = 30 \text{ kPa}$

Assume that the $\rm H_2$ generation rate was measured to be

 $\begin{array}{rcl} H_{M} &=& 0.2 \pm 0.1 \mbox{ gmole } H_{2}/\mbox{Hr at} \\ T_{M} &=& 348 \pm 2 \ ^{\circ}\mbox{K and} \\ P_{M} &=& 10 \pm 0.1 \mbox{ kPa water vapor} \end{array}$

Using the equations in Section B.1, the following can be calculated:

$$\begin{split} R_{\rm H} &= 23.8 \text{ g U/Hr}, \ \Delta R_{\rm H} = 11.9 \text{ g U/Hr} \\ R_{\rm LIMIT} &= 28.9 \text{ g U/Hr} \\ \partial R_{\rm LIMIT} / \partial P_{\rm H} &= 1.44 \text{ g U/Hr/kPa}, \ \partial R_{\rm LIMIT} / \partial T_{\rm H} = 1.18 \text{ g U/Hr/}^{\circ} \text{K} \\ \Delta R_{\rm LIMIT} &= 2.36 \text{ g U/Hr} \\ \text{f} &= 0.82 \\ \partial f / \partial R_{\rm H} &= 0.0035, \ \partial f / \partial R_{\rm LIMIT} = -0.0286, \ \Delta f = 0.11 \end{split}$$

Therefore,

$$f = 0.82 \pm 0.11 \le 1$$

APPENDIX C

DENSITY DETERMINATION METHOD OF ESTIMATING MCO CORROSION PRODUCT INVENTORY

C.1 INTRODUCTION

This appendix explores the viability of using fuel assembly density differences to quantify residual corrosion products included in an MCO. It concludes that the sensitivity of instruments required would be quire high for accurate determinations; to identify 1%, or 0.2 Kg of corrosion product on a 20 Kg fuel assembly requires an instrument sensitive to better than 0.005% (1 gram) differential mass. This sensitivity would allow identification of 50 Kg of corrosion products in a 5000 Kg MCO load. Instruments with this accuracy are not available for production applications.

Alternatively, with credible instrumentation, it may be possible to measure corrosion product against a 300 Kg limit. Not that the uncertainty of the initial density for an assembly will dominate the accuracy of this measurement.

C.2 BASIC CONCEPT

Table C-1 provides a summary of the steps anticipated to be used in implementation of this measurement concept. A fuel assembly, in the basin, would be weighed while submerged. It would then be enclosed in a bell, which would be blown dry. The dry weight of the assembly would be determined. By comparing the wet and dry mass of an assembly, the density can be obtained.

Corrosion products have a lower density than uranium. Reduction in density below the as-built assembly is a clear indication of corrosion product accumulation.

C.3 BACKGROUND

The fuel is composed of a range of element sizes. Table C-2 provides a survey of some parameters used in the following analysis.

Note that the effective assembly density decreases for shorter elements. The density ranges from 16.7 to 16.9 g/cm^3 for a total assembly. The inner element of a type C assembly has an anticipated density of 16.6 g/cm^3 , while an outer element of type E has a density of 16.9 g/cm^3 .

Table C-1. Gravimetric Corrosion Product Identification Procedure

Step	Action
1.	Identify assembly type
2.	Weigh assembly
3.	Compare weight with expected fabrication weight.
ļ	a) If weight exceeds anticipated weight by more than 0.5 Kg, go to step 4.
	b) If weight is within 0.5 Kg of anticipated weight, go to step 6.
	c) If weight is less than anticipated weight by more than 0.5 Kg, go to step 5.
4.	Rework assembly through cleaning process to remove corrosion product
5.	Assess density of element
	a) If density signature exceeds 20% corrosion product threshold, go to step 4.
	b) Otherwise go to step 6.
6.	Place assembly in fuel or scrap basket

Table C-2. Mark IV Fuel Assembly Parameters

Type -	Clad Mass, Kg		Fuel Mass, Kg		Total Mass, Kg	Effective		
Length Code	Inner	Outer	Inner	Outer		Assembly Density, g/cm ³		
E	0.55	1.09	7.48	16.0	25.1	16.90		
S	0.52	1.04	7.03	15.0	23.6	16.88		
A	0.50	0.99	6.62	14.1	22.2	16.85		
С	0.40	0.79	4.94	10.5	16.6	16.73		

Reference: Willis, 1995

With corrosion, the mass of an assembly will rise, assuming no loss of corrosion products. Table C-3 presents the fraction mass increase for corrosion products over the base metal.

Table L-3. Fractional Mass Lhange for Corrosion Produ	Produc	rrosion	TOR	unange	Mass	Ionai	Fract	l-3.	ladie
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Corrosion Product	Fractional Mass Change					
UO ₂	0.13					
UO ₃	0.20					
UO3 • 2 H ₂ O	0.35					

Hence a total assembly mass increase of 0.5 Kg with no sign of fuel loss could consist of up to 17% UO, by mass, although volume arguments will show that this is not credible. Hence, a <u>high</u> mass is unlikely to be detected.

The density of corrosion products, in particle form, is less than 4 g/cm_3 . Hence, the uranium will undergo a volume growth of a factor of 4.75 or more. Corrosion of more than 1% of an assembly without detectable external damage is unlikely.

C.4 EVALUATION OF DENSITY AND MEASUREMENT UNCERTAINTY

C.4.1 Corrosion product Estimate Equations

Let:

- M_{wet} = The immersed mass of an assembly (measured by weight measurement instrument)
- M_{drv} = The dry mass of the assembly
- ρ_{water} = Density of water (assumed to be 1 g/cm³)
- V = The volume of the assembly

 ρ = The measured density of the assembly

$$V = (M_{dry} - M_{wet}) / \rho_{water} = M_{dry} - M_{wet}$$

$$\rho = M_{dry}/V = M_{dry}/(M_{dry} - M_{wet})$$

Using a simplified assumption that the density of the intact portions of an assembly does not change appreciably during corrosion, let:

x = Fraction of corroded material by volume

 $\rho_{\rm u}$ = The assembly density = 16.8 g/cm³

 ρ_c = The corrosion product theoretical density

Then,

 $\rho = (1 - x) \rho_u + x \rho_c$

Now let the fraction mass change be f.

 $f = (M_{dry} - M_{wet})/M_{dry} = 1/\rho$

Hence,

 $\rho = 1/f = (1 - x) \rho_{\mu} + x \rho_{c}$

Solving for x

or
$$x = (\rho_u - \rho) / (\rho_u - \rho_c)$$
$$x = (16.8 - \rho) / (16.8 - 10.97)$$

Note that ρ ranges from 16.8 to 10.97. Table C-4 presents the relationships between measured density and the percentage of corrosion products in the sample. Also note that 10.97 is selected as the maximum theoretical density of UO₂, which is the most dense of the corrosion products. Actual corrosion products such as UO₃ with lower densities will yield higher than actual (conservative) corrosion product fractions based on this relationship.

Table C-4. Measured Density and Corrosion Product Quantity

Measured	Percent
Density,	Corrosion
g/cm³	Product
16.8	0.0
16.7	1.7
16.6	3.4
16.5	5.1
16.4	6.9
16.3	8.6
16.2	10.3
16.1	12.0
16.0	13.7
15.2	27.4
14.4	41.2
13.6	54.9
12.8	68.6
12.0	82.3
11.2	90.1

The practicality of measuring these densities with the sensitivity required to achieve 1% accuracy can be seen by relating to the fractional mass change as a function of density. Table C-5 shows the instrument accuracy required to perform the measurement with resolution sufficient to quantify corrosion products as a function of minimum percentage corrosion product resolution.

Percentage Corrosion Products in Sample	Predicted Density, g/cm ⁵	Fractional Mass Change between Wet and Dry	Mass Change for a 20 Kg Assembly Mass, Kg		
0 1 2 3 4 5 6 7 8	16.80 16.74 16.68 16.62 16.57 16.51 16.45 16.39 16.33 16.27	0.0595 0.0597 0.0599 0.0602 0.0604 0.0606 0.0608 0.0608 0.0610 0.0612 0.0612	1.190 1.195 1.199 1.203 1.207 1.212 1.216 1.220 1.225 1.225		
10	16.21	0.0617	1.233		

Table C-5. Variation of Assembly Mass with Residual Corrosion Product

To resolve a 1% difference in corrosion product quantity, one must measure the mass difference to within 5 grams, requiring a measurement system accurate to better than 0.005%. This level of accuracy in a production instrument is not easily achieved.

C.5 OTHER ERRORS

The preceding analyses did not factor in variability in the initial fuel assembly density resulting from production uncertainties such as clad quantity and fuel quantity. Further, corrosion has been shown to break off portions of the clad, adding to a term not considered in the above analyses. A decision to pursue density measurements would require thoughtful evaluation of such effects.

C.6 CONCLUSION

To achieve a 50 Kg MCO corrosion product limit, the corrosion product quantity per assembly would have to be less than 1% of each assembly's mass. This would require an instrument sensitive to better than 0.005% (1 gram) differential mass for a 20 Kg assembly. Since the original density variation of an assembly were likely to have exceeded the limit required for 1% resolution, this method is not viable as a production technique, and may not be viable overall, regardless of the instrument sensitivity.

Evaluating the problem alternately, one can assess what production instruments can reasonably achieve for continuous operation. A catalog survey indicates that 0.5% accuracy and 0.01% resolution is achievable. If an instrument resolution to 0.01% were used, this would allow identification of a 10% volume corrosion fraction or roughly 6% mass fraction. Hence, for a 5000 Kg MCO fuel loading, 300 Kg of corrosion product could be measured with some confidence.

APPENDIX D

MCO CORROSION PRODUCT INVENTORY ESTIMATE BASED ON FUEL BASKET MASS

D.1 CONCEPT DESCRIPTION

Because oxidized fuel may be trapped inside the cladding of apparently intact and clean fuel assembly, a method of established an upper bound for corrosion product inside fuel in needed. The proposed method consist of calculating the maximum weight of an intact element and comparing this weight to the minimum measured weight after cleaning. The difference would be assumed to be the change in weight due to conversion of uranium with a specific gravity of 18.75 g/cc to oxide with a density of 4 g/cc. A basket of fuel could be weighted under water before placement in the MCO. The underwater weighing should not significantly reduce the accuracy of the weight. The uranium fuel with cladding is about 15 times as dense as water and the density of the water will be very predictable thus an accurate weight can be determined.

D.2 CORROSION PRODUCT INVENTORY EVALUATION

The corrosion product inventory estimate is based on a comparison of the measured mass of fuel in a basket which is compared to the theoretical mass of the fuel elements. Development of the equations is as follows:

Let:

Theoretical total mass of fuel in a basket M. =

 M_{u} = Theoretical mass of uranium in a basket M_{e} = Theoretical mass of cladding in a basket

Theoretical mass of cladding in a basket

 $M_{*} = M_{*} + M_{*}$

For the measured mass of fuel, let:

- M_b = The measured total mass of fuel in a weighed basket
- U^D= The mass of uranium in a weighed basket
- C = The mass of cladding in a weighed basket
- S = The mass of corrosion product in a weighed basket

 $M_{\rm b} = U + C + S$

Assume that any difference in the measured and theoretical mass of fuel in the basket is due to the presence of corrosion product, such that

 $M_{c} = C$

Assume that the volume of fuel/corrosion product enclosed by cladding in the weighed basket is bounded by the theoretical fuel assembly dimensions (e.g., no swollen cladding regions are present in fuel within the weighed basket), then for:

 $\rho_{\rm u} =$ The density of uranium metal $\rho_{\rm s} =$ The density of corrosion product

$$U/\rho_u + S/\rho_s \le (M_t - M_c)/\rho_u$$

which can be rearranged

 $U + S(\rho_u/\rho_e) \leq (M_t - M_e)$

Noting that $U = M_h - C - S$, $M_h = C$, and solving for S yields:

 $S \leq (M_{r} - M_{b})/(\rho_{u}/\rho_{s} - 1)$

Figure D-1 provides a plot of the maximum corrosion product inventory estimated in a fuel basket as a function of the weighed fuel mass assuming all fuel assemblies are E length Mk IV assemblies. Similar plots could be developed for the different fuel lengths or a mix of fuel lengths to implement this approach.

D.3 ESTIMATE UNCERTAINTIES

For this approach to work several conditions must be met:

- Fuel assemblies must be clean and free of surface corrosion product
- Assemblies must not be swollen by corrosion under the cladding
- Length and model of each assembly must be known
- Accurate tare and loaded weights for each basket in an MCO must be available

Given these constraints, the limit of detection for corrosion product will be a function of the uncertainty in fabrication dimensions and the net weight of the basket. Although the actual fabrication tolerances and the weighing precision is not available, a reasonable set of assumptions can be made for illustration.

fabrication +/- 0.2% of diameter and length weight +/- 0.15% of net for both tare and gross

The two sigma difference in weight is $2^{(0.2^2 + 0.4^2 + 0.15^2 + 0.15^2)} = \frac{0.99\%}{1000}$

The total weight of fuel is 6340 kg per MCO thus the two sigma uncertainty is $\underline{64 \ \text{kg}}$

The conversion of this volume of uranium to corrosion product is the uranium weight divided by the difference in density between corrosion product and metal and multiplied by the density of corrosion product.

64 kg * 4 g/cc / (18.74 g/cc - 4 g/cc) = <u>17.4 kg of corrosion product/MCO</u>

These could be optimistic assumed values and the actual uncertainty could be greater but, this a good first estimate of the ability of this approach to establishing a maximum corrosion product limit for clean fuel.





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APPENDIX E

MCO HYDROGEN INVENTORY ESTIMATE BASED ON ISOTOPIC EXCHANGE

E.1 CONCEPT DESCRIPTION

The hydrogen inventory within a MCO can theoretically be measured by introducing a known quantity of hydrogen isotope (either deuterium or tritium) into the MCO vapor space. The method then relies on isotopic exchange occurring between all hydrogen isotopes incorporated in compounds within the MCO until equilibrium is achieved. The steps associated with implementing this alternative are:

- Measure the void volume (or gas vapor space volume) of the MCO, partial pressure of water in the void space, and partial pressure of hydrogen in the void space.
- Measure the ratio of tritium (or deuterium) atoms to total hydrogen atoms of gas in the vapor space (e.g., mass spectroscopy analysis of gas sample).
- 3. Introduce a known quantity of tritium into the MCO vapor space.
- 4. Wait a sufficient time period for isotopic equilibrium to be achieved.
- 5. Measure the ratio of tritium atoms to total hydrogen atoms in the gas vapor space a second time.

E.2 EVALUATION OF HYDROGEN INVENTORY

The development of the method for calculating the MCO hydrogen inventory from an isotope spike addition is as follows:

Let:

 H_v = Total hydrogen atoms in MCO vapor space, gmole H

 $V_{void} = Vapor space of MCO, L$

- T = MCO gas temperature, °K
- P_{u} = Partial pressure of water measured in the MCO vapor space, atm
- P_{μ} = Partial pressure of hydrogen measured in the MCO vapor space, atm
- γ = Ratio of tritium (or deuterium) to total hydrogen, gmole ³H/gmole H
- A = Quantity of tritium in spike addition, gmole 3 H
- H_s = Total hydrogen atoms in MCO solids/liquids, gmole H
- y = Fraction of tritium spike remaining in vapor phase when isotopic equilibrium achieved

At initial conditions:

$$H_{vi} = 2 (P_{\mu} V_{void})/RT + 2 (P_{\mu} V_{void})/RT$$

Assuming at isotopic equilibrium that γ in the vapor space is equal to γ in the solids/liquids remaining in the MCO, a balance for the tritium spike addition yields:

In: $H_{vi} \gamma_i + A + H_{si} \gamma_i$

Out: $(H_{vi} + y A) \gamma_f + (H_{si} + (1-y) A) \gamma_f$

Accumulation: 0

This can be solved for the total hydrogen atoms in MCO solids/liquids, H.,.

$$H_{si} = [A (1 - \gamma_f) - H_{vi} (\gamma_f - \gamma_i)]/(\gamma_f - \gamma_i)$$

Note that this calculates the total hydrogen, not the water or corrosion product inventory. Water can be estimated by assuming all the hydrogen measured in solids is as water. This assumption would also be equivalent to assuming that the hydrogen measured reflects the impact of surface absorbed water, while other hydrogen is tied up as uranium hydride in the bulk solid matrix of materials and is less accessible for isotopic exchange over practical time periods. The corrosion product inventory would have to be inferred from the water estimate using either an assumed moisture fraction or specific surface area for particulate.

E.3 EXAMPLE CALCULATION

An example calculation is presented which may be similar to application of this technique at the conclusion of Hot Conditioning System activities. Assume a tritium spike is used in the measurement technique for a MCO at initial conditions of 373 °K, vapor space of 500 L, and measured partial pressure of water at 0.01 atm. Assume the initial partial pressure of hydrogen and ratio of tritium to hydrogen measured in the gas vapor space are negligible. Assume a spike addition of 1 g tritium gas (0.167 gmole) is added to the vapor space and allowed to come to equilibrium. Based on this example, $H_{\rm yi} = 0.33$ gmole total hydrogen at the initial conditions. Table E-1 presents the resultant hydrogen and water inventory estimated for an MCO for a range of assumed measured tritium to hydrogen ratios after isotopic equilibrium is achieved. This indicates that measurement of tritium to total hydrogen ratios in the gas space must be demonstrated to be on the order of 0.001 in order to indicate that the MCO water inventory is within constraints for sealed

Measured	Total Hydrogen	Total MCO Water
Tritium/Hydrogen Ratio	Inventory in	Inventory in
at Equilibrium, y _f	Solids, gmole	Solids, Kg
0.1	1.17	0.011
0.01	16.2	0.15
0.001	167.	1.5
0.0001	1670.	15.

Table E-1. Example Calculation Results of Isotopic Exchange Method

DISTRIBUTION SHEET								
То	From				Pa	age 1 of 1		
Distribution	Proces	rocess Engineering			Date 7/17/96			
Project Title/Work Order				EDT No. 616110				
MCO Pressurization Monitoring an Spent Nuclear Fuel Project	ıd Contr	Control Methodology for the				ECN No. N/A		
Name		MSIN	Text With All Attach.	Text Onl	y	Attach./ Appendix Only	EDT/ECN Only	
<pre>G. D. Bazinet K. H. Bergsman F. W. Bradshaw B. S. Carlisie R. G. Cowan G. E. Culley D. R. Duncan J. R. Frederickson W. D. Gallo L. J. Garvin III E. W. Gerber L. H. Goldmann J. J. Irwin A. T. Kee M. K. Mahaffey D. W. McNally T. D. Merkling C. R. Miska R. P. Omberg J. W. Osborne A. L. Pajunen (15) P. A. Scott E. J. Shen K. E. Smith J. A. Swenson J. C. Wiborg </pre>		H5-27 R3-85 R3-85 R3-86 R3-86 R3-86 R3-86 R3-85 R3-85 R3-86 R3-85 R3-85 G1-11 B1-10 R3-86 R3-85 X3-80 R3-85 X3-80 R3-87 R3-86 R3-87 R3-86 R3-87 R3-86 R3-87 R3-86 R3-87 R3-86 R3-87 R3-86 R3-87 R3-86 R3-87 R3-86 R3-87 R3-86 R3-87 R3-86 R3-87 R3-86 R3-87 R3-86 R3-87 R3-86 R3-87 R3-86	***					
proj giles		R3-11	X					