Idaho
National
Engineering
Laboratory

INEL-95/00437

INEL--95/00437

June 16-20 1996

CONF-9606116--44

Drying Studies of Simulated DOE Aluminum Plate Fuels

Rebecca E. Lords
Williams E. Windes
John C. Crepeau
Rod W. Sidwell

Presented at
American Nuclear Society
Topical Meeting on
DOE Spent Nuclear Fuel and
Fissile Material Management

Lockheed
Idaho Technologies Company

DRYING STUDIES OF SIMULATED DOE ALUMINUM PLATE FUELS

Rebecca E. Lords
William E. Windes
INEL Spent Nuclear Fuel Program
Lockheed Martin Idaho Technologies
P.O. Box 1625
Idaho Falls, ID 83415-3114
208-526-3874

John C. Crepeau
Rod W. Sidwell
University of Idaho
Department of Mechanical Engineering
P.O. Box 50778
Idaho Falls, ID 83405
208-526-1388

ABSTRACT

Experiments have been conducted to validate the Idaho National Engineering Laboratory (INEL) drying procedures for preparation of corroded aluminum plate fuel for dry storage in an existing vented (and filtered) fuel storage facility. A mixture of hydrated aluminum oxide bound with a clay was used to model the aluminum corrosion product and sediment expected in these Department of Energy (DOE) owned fuel types. Previous studies demonstrated that the current drying procedures are adequate for removal of free water inside the storage canister and for transfer of this fuel to a vented dry storage facility. However, using these same drying procedures, the simulated corrosion product was found to be difficult to dry completely from between the aluminum clad plates of the fuel.

Another related set of experiments was designed to ensure that the fuel would not be damaged during the drying process. Aluminum plate fuels are susceptible to pitting damage on the cladding that can result in a portion of UAl_x fuel meat being disgorged. This would leave a water-filled void beneath the pit in the cladding. The question was whether bursting would occur when water in the void flashes to steam, causing separation of the cladding from the fuel, and/or possible rupture. Aluminum coupons were fabricated to model damaged fuel plates. These coupons do not rupture or sustain any visible damage during credible drying scenarios.

I. INTRODUCTION

The Idaho National Engineering Laboratory (INEL) currently stores a wide variety of spent nuclear fuel. The fuel was originally intended to be stored underwater for a short period of thermal cooling, then removed and reprocessed. However, it has been stored underwater for much longer than originally anticipated. During this time dust and airborne desert soil have entered the oldest INEL pool (ICPP-603), accumulating on the fuel. Also, the aluminum fuel cladding has corroded compromising the exposed surfaces of the fuel. Plans are now underway to move some of the more vulnerable aluminum plate type fuels into dry storage.

In preparation for dry storage of the fuel a drying and canning station is being built at the INEL. Experiments to validate the drying procedures to be used in this drying station are ongoing. Initially, there were three experimental objectives specifically designed to support drying operations: to determine the influence of corrosion products on the drying process, establish temperature distribution inside the canister during heating, and evaluate the potential for fuel damage during drying.

Residual moisture contained within hydrated corrosion products between the fuel plates may be difficult to remove during normal drying practices due to the tenacity of the chemically bound waters of hydration. Any remaining moisture may lead to additional degradation of the fuel and canister materials and eventual build up of excess pressure in a sealed storage system.

Experimental determination of the temperature distribution throughout the vessel and the mock fuel during heating was needed. Accurate measurements of the fuel element and canister temperatures are necessary to ensure appropriate drying conditions. Practical considerations restrict the ability to monitor the canister contents. The data from these experiments provide a baseline reference for projection of fuel temperature during the drying process based on the available vessel air temperature.

Concern for possible bursting problems arose from visual inspections of the fuel elements indicating extensive pitting corrosion on the external cladding of the outer fuel plates. The UAl_x fuel matrix between the aluminum cladding may have been disgorged with water filling the subsequent voids beneath the pitted surface.[1, 2] During aggressive drying procedures additional damage to the thinly clad fuel plates was anticipated due to the rapid exit of water from any existing voids.

II. EXPERIMENTAL SET-UP

A full-size mock-up of the drying and canning station has been designed, built and tested at the INEL to perform experiments on drying mock aluminum plate fuel elements. A number of mock Advanced Test Reactor (ATR) fuel elements, representative of the aluminum plate types, have been constructed for this work. Figure 1 shows a schematic diagram of the drying station along with a sketch of the mock ATR fuel elements.

The drying chamber is fabricated from an 18 inch, schedule 40 stainless steel pipe approximately

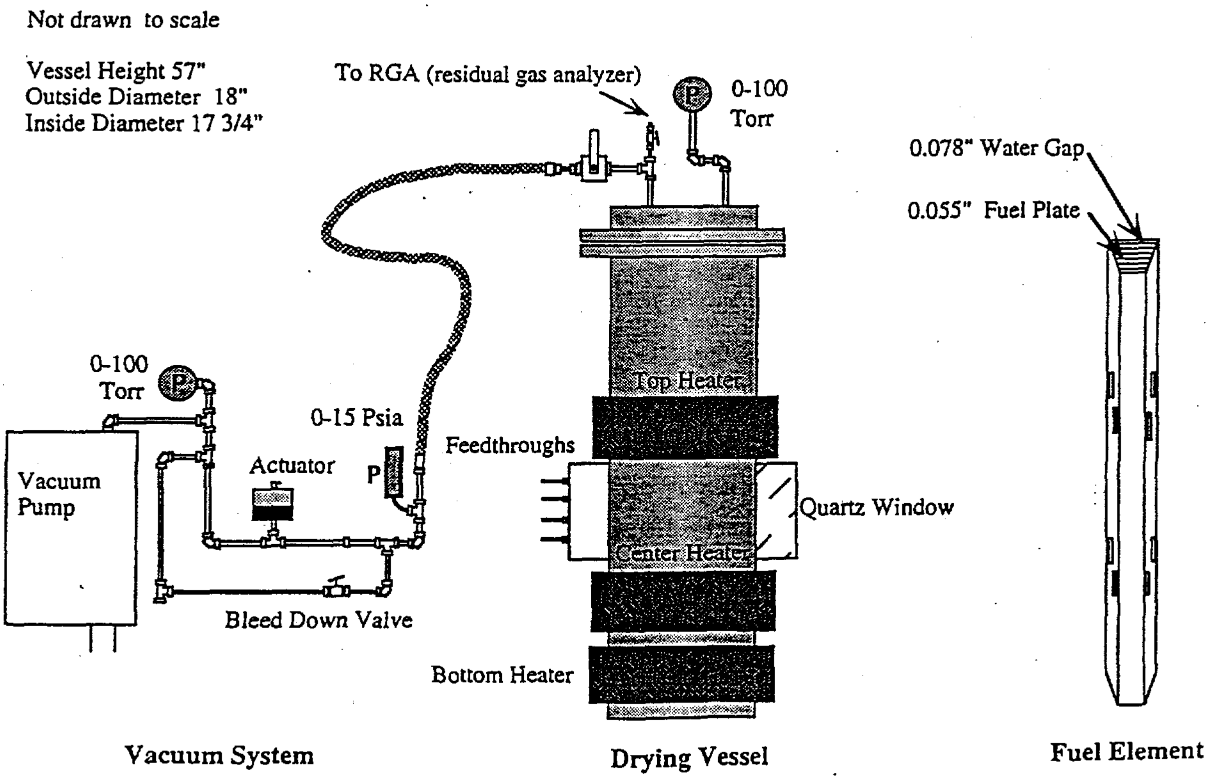


Figure 1. Left: Mock Drying Station; Right: ATR Fuel Element

57 inches tall. A companion chamber is available to accommodate longer samples in a stacked chamber configuration. Three heating bands on the exterior of each chamber enable strict control of the temperature and heating rate while a vacuum pump can attain chamber pressures down to ~0.5 torr. Two side ports are installed in the chamber mock-up to allow visual confirmation of interior status through a quartz window and to provide access for the instrumentation leads. Thermocouples, pressure transducers and heat flux monitors are available to record data from each experiment.

III. RESULTS

A. Simulated Corrosion Product

Since the predominant corrosion product is expected to be a hydrated aluminum oxide, initial tests were intended to determine if the drying station could remove a measured amount of water that is physically mixed with a simulated corrosion product. The mixing allowed the majority of the water to be physically, not chemically, bound to the exterior surface of the corrosion product particles.

For simplicity, initial tests were conducted using commercially available pseudo-boehmite powder, $Al_2O_3 \cdot 1.3H_2O$. Drying this pseudo-boehmite resulted in powder being lost in the vacuum system and chamber. The aluminum oxide particulate was so fine that it was driven out of the sample dish by boiling, modest pressure decreases, and even through sublimation. Consequently, dust was spread throughout the chamber and vacuum system making correlations between weight loss and remaining moisture levels difficult.

Further observation of the ICPP-603 fuel storage basin environment and pool sludge indicated that actual material on the fuel will be a combination of components rather than pure aluminum corrosion product. Elemental analysis of corrosion on the aluminum coupon exposed to ICPP-603 basin water supports this assessment.[3, 4] Airborne dust and silt is blown into the ICPP-603 facility from the desert, and this sediment drifts into the open cooling channels in the fuel forming a mixed deposit of sediment and aluminum oxide.

A mixture of clays, aluminum oxide and water was determined to be a more accurate representation of the actual deposit expected in the plate fuels. This mixture also creates a product that has improved experimental properties with the clay acting as a binder for the aluminum oxide powder avoiding loss to the system as mentioned previously. This mixture was developed to parallel the INEL soil analysis with measured amounts of pseudo-boehmite added.[5, 6, 7] Table 1 shows the nominal weight percent of the materials used in the simulated corrosion product mixture. Note that commercially available bentonite was used in place of montmorillonite, the dominant constituent in INEL soils. Bentonite is composed mostly of montmorillonite and typically includes some of the illite and kaolinite also present in soils at the INEL. The water was blended with the dry mixture to achieve a thin putty. This putty was spread between the plates of the mock fuel elements, and the entire assembly was subjected to a series of temperature ranges, heating rates, and pressure levels to support INEL drying parameters.

Table 1.
Composition of Simulated Corrosion Product Mixture.

Nominal Weight %	Dry	Wet
Bentonite	31	11
Kaolinite	27	10
Pseudo-boehmite	42	15
Water	—	64

B. Vacuum Pressures and Drying Procedure

The current drying procedure at the INEL is limited by time constraints and safe handling practices. The proposed drying procedure consists of two distinct parts; vacuum drying to remove the free water in the canister and high temperature bake out to ensure all the free water has been removed from the system. Previous work has shown that for relatively simple fuel configurations with little or no corrosion product on the exposed surfaces, vacuum drying will remove all free water in the canister.[8, 9]

The preferred means of remotely determining if the free water has been removed is to monitor the pressure inside the chamber. Once the pressure has dropped below 3 torr (and stays at this same pressure after isolation from the pump) the canister can be assumed to be dry.[10] However, for fuels containing hydrated material (i.e. hydrated aluminum corrosion product) the moisture is only gradually liberated during the vacuum drying process. This reaction is very slow at ambient temperature, but enough moisture remains physically or chemically bound to the hydrated material to obscure the vacuum drying endpoint.

This work demonstrates that vacuum drying does not remove bound water in the simulated corrosion product between the plates in aluminum clad plate fuels. Table 2 shows data for drying a simulated corroded aluminum plate fuel element under several variations of proposed drying procedures. Depending upon the length of time under vacuum, a greater or lesser amount of moisture remains within the simulated corrosion product. Note that the first 16-17 percent of the estimated water removed is driven off with minimal vacuum time. Additional vacuum time only removes up to about 28 percent of the estimated water present; continuing to apply vacuum beyond four hours does not achieve significantly more water removal. The two hour bake period after the vacuum step drives off more of the water. To date, the maximum length of time spent vacuum drying one element to a qualitative state of dryness (based on visual appearance) is about three days.

C. Heating Rates and Temperature Ranges

After satisfactory vacuum drying, the chamber is vented and heated to a nominal inside air temperature of 100°C. This temperature is held for a period of two hours to drive off any residual water.

Physical and operational limitations of the drying station do not allow for thermocouples to be directly attached to any of the fuel elements. This 100°C inside air temperature will be measured by

Table 2. Averaged Results from Various Drying Procedures.

V = Vacuum time H = Heating time	V = 0.5 hr H = 0 hr	V = 2 hr H = 0 hr	V = 4 hr H = 0 hr	V = 6 hr H = 0 hr	V = 2 hr H = 2 hr	V = 4 hr H = 2 hr	V = 6 hr H = 2 hr *
Weight percent change of simulant after drying, %	10%	11%	18%	18%	29%	39%	40%
Estimated total water in simulant, g	414	444	387	405	405	389	335
Estimated water removed from simulant, g	66	77	108	114	183	239	208
Estimated weight percent of water removed from simulant after drying, %	16%	17%	28%	28%	45%	61%	62%

* Only 1 test run for these conditions

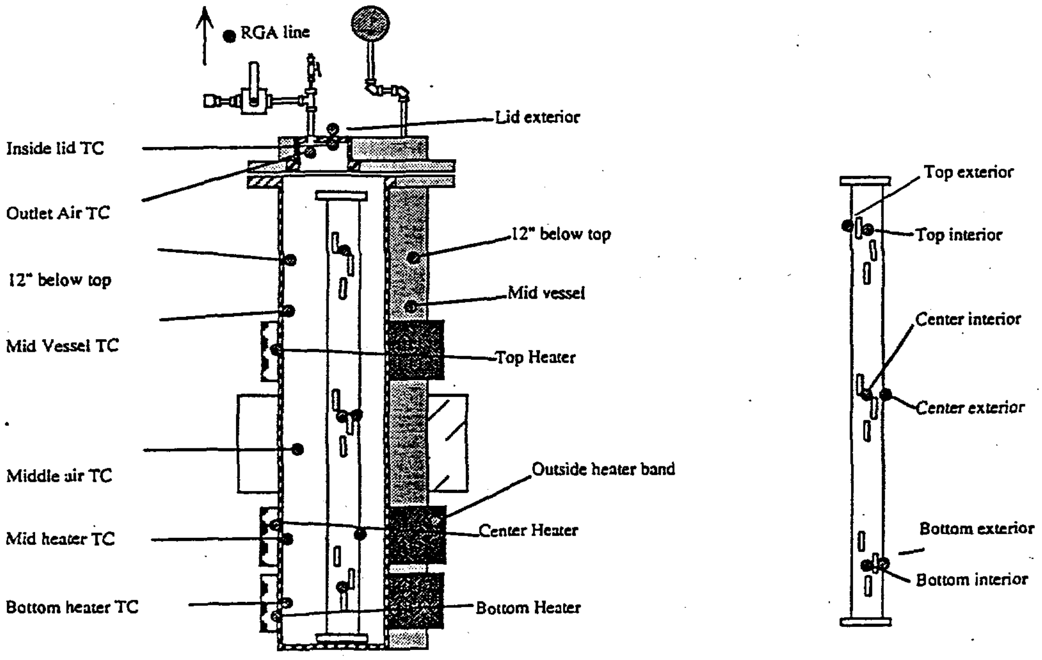
thermocouples located in the top portion of the canister. Projections must be made to ensure that the fuel elements actually reach the desired operating temperature for the bake out period. Temperatures throughout the drying station mock-up and various locations on the mock fuel have been monitored for different bake-out temperatures to provide a baseline. Figure 2 shows all of the thermocouple locations used in the mock-up.

Inside canister air and mock fuel element temperatures at several locations are presented for several potential operating temperatures in Figure 3. These are typical temperature profiles that are expected in the actual drying station during the bake-out. Note that for the nominal vessel inside air temperature of 100°C the exterior mock fuel element temperature reached 100°C and the center interior of the mock fuel element also reached a temperature of 100°C after two hours. Note in Figure 4 that when the simulated corrosion product is in place for the same nominal vessel interior air temperature of 100°C the exterior mock corroded fuel element temperature only reached 95°C and the center interior of the mock corroded fuel element only reached a temperature of 90°C after the same two hour period.

D. Potential Damage to Fuel During Drying

Aluminum coupons were fabricated to simulate voids within the fuel plates to help determine the bursting characteristics of the fuel plates during drying. The intent was to establish a safe temperature and pressure limit for the drying protocol to ensure that the plate fuels are not damaged during the drying operations.

The coupons have small cavities (a 0.79 inch diameter hole, 0.015 inch deep) machined into a solid aluminum plate 0.035 inch thick to imitate the cavities expected in the actual fuel elements. A 0.006 inch thick aluminum foil (the minimum cladding thickness allowable by manufacturing specifications for ATR fuel plates) was attached by epoxy over the machined cavity in the coupon to simulate the cladding, and water was then injected into the void. This configuration adequately models a water pocket trapped in an aluminum spent fuel plate before drying occurs. Various heating rates and pressure changes inside the canister were employed to establish conditions which could result in rupture of the foil.



Drying Vessel

ATR Fuel Elelment

Figure 2. Thermocouple Locations In Drying Vessel And Mock Element.

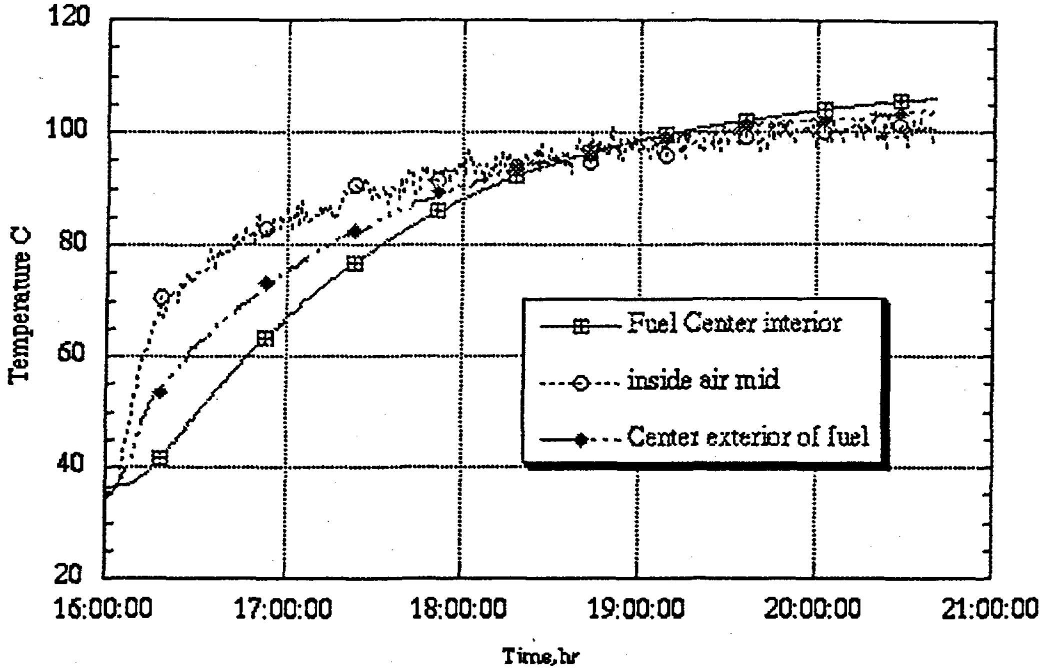


Figure 3. Temperature Inside Aluminum Plate Fuel Without Corrosion Product.

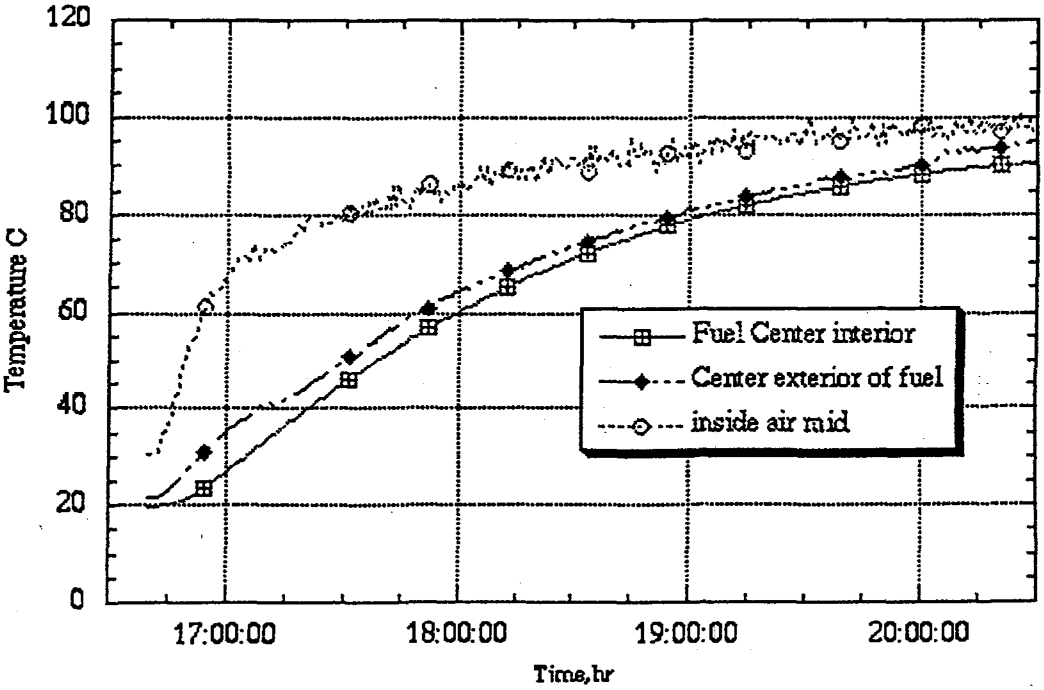


Figure 4. Temperature Inside Aluminum Plate Fuel Element With Corrosion Product.

None of the proposed drying temperature and vacuum combinations caused any deformation or damage to the coupons. Observations indicated that the liquid water was rapidly expelled from the cavity through the breach. Even the most extreme combination of conditions, where the coupon was heated first to 93°C and then exposed to a sudden pressure drop, did not cause visible damage to the coupon.

IV. CONCLUSIONS

Clays seem to be effective additives for simulating the combination of materials expected to be associated with the fuel after extended periods of underwater storage at ICPP-603. They improve the level of confidence in the mock-up tests by providing a hydrated mixture which bounds the corrosion product drying problem. The clays provide a binder which mitigates the previously observed difficulty of mass loss due to disturbance of dust particulate from the pure pseudo-boehmite simulated aluminum corrosion product. The clay mixture also adheres to the mock fuel plates better than the pseudo-boehmite alone.

The current drying procedures are adequate to remove the free water from the fuel canisters for eventual transfer to a vented storage facility. However, the current drying procedures are not adequate to remove bound water from hydrated corrosion products inside the fuel elements. A more aggressive drying procedure must be established for the corroded fuel.

The temperature in pristine fuel elements can be expected to closely follow the inside air temperature within the drying station (neglecting any heat from the radioactive fuel components). If the fuel channels contain corrosion product and sediment, the temperature in the fuel element may lag behind the temperature within the drying chamber by as much as 10°C. If the process objective is to hold the fuel at a specific temperature, this lag should be accounted for in the operating procedure.

Rupture of cladding or damage to the fuel due to occluded water flashing to steam appears not to be an issue. The aluminum coupon tests established that the geometry and volume of water pockets anticipated would not lead to damage to the fuel under any combination of the proposed operating parameters.

V. FUTURE WORK

Results from this study indicate that the current INEL drying procedure can be improved for corroded aluminum plate fuels. The simulated hydrated corrosion product material will be analyzed to determine the quantity of moisture remaining after the drying process. An optimized drying procedure using vacuum and heating processes in combination will be established.

A more reliable means of remotely determining the moisture level inside the canister must be established. The addition of a Residual Gas Analyzer (RGA) to the exhaust piping will be explored as an option to provide real time monitoring of the exit gas during drying. Correlation of RGA data to the optimized drying procedure may be used to indicate moisture content throughout the drying process. The RGA may also be used to detect other gases released during drying.

The true temperature profiles of the elements and canister during drying will be influenced by the decay heat generated by the radionuclides in the spent fuel. By artificially heating the mock elements within the canister, the temperature distribution will be monitored to generate a refined temperature profile for the canister and contents during the bake-out. This data can be applied to the drying process and eventually to the fuel storage environment.

ACKNOWLEDGEMENTS

Work supported by the U. S. Department of Energy, Assistant Secretary for Environmental Management, under DOE Idaho Operations Office Contract DE-AC07-94ID13223.

REFERENCES

1. K. Vinjamuri, *Postirradiation Examination of Advanced Test Reactor Fuel Elements XA377N and XA379N*, EGG-TFBP-5968, EG&G Idaho, Inc., Idaho Falls, ID, September 1982.

2. S. L. Seiffert and A. H. Novick, *Destructive Examination of Defective Fuel Plates From ATR Fuel Elements A197F and XA456N, PR-T-79-016*, EG&G Idaho, Inc., Idaho Falls, ID, August 1979.

3. P. A. Anderson, "Analysis of corrosion product on 6061 T-6 aluminum alloy coupon exposed to CPP-603 basin water", PAA-13-94 to C. V. Shelton-Davis, Idaho National Engineering Laboratory, June 23, 1994.

4. P. A. Anderson, "Characterization of Corrosion Solids on ATR Fuel Stored in the CPP-603 Basin", PAA-03-95 to C. V. Shelton-Davis, Idaho National Engineering Laboratory, November 6, 1995.

5. R. L. Nace, M. Deutch, and P. T. Voegeli, *Geography, Geology, and Water Resources of the National Reactor Testing Station, Idaho: Part 2. Geography and Geology*, pp. 141-152, U. S. Geological Survey, Boise, ID (1956).

6. C. S. Hurlbut, Jr., Dana's Manual of Mineralogy, Sixteenth Edition, pp. 518-530 John Wiley & Sons, Inc., New York, NY (1952).

7. R. E. Grim, Clay Mineralogy, International Series in the Earth Sciences, pp. 29-38, 361-373, McGraw-Hill Book Company, Inc., New York, NY (1953).

8. A. H. George, J. Rogers, and J. Fleming, *Final Report - Vacuum Drying of Fuel Containers*, Montana State University, prepared for D. L. Peters, Westinghouse Idaho Nuclear Company, Performance Period 1 June 1993 to 14 January 1994.

9. J. O'Brien, D. Phetteplace, M. Sprenger, and H. Welland, *ICPP-603 Fuel Canning Project May Street South Vacuum Drying Mockup Station Test Report Document*, EG&G Project File No. 015635, Idaho National Engineering Laboratory, September 1994.

10. *Safety Analysis Report for the Standardized NUHOMS® Horizontal Modular Storage System for Irradiated Nuclear Fuel*, Volume I Nonproprietary Report, NUH-003 Revision 1 NUH003.0103, Pacific Nuclear Fuel Services, Inc., San Jose, CA, September 1991.