Pacific Northwest National Laboratory

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Spent Fuel Drying System Test Results (Second Dry-Run)

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July 1998

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Pacific Northwest National Laboratory
Richland, Washington 99352

Summary

The test described in this report is part of a series of tests being conducted by Pacific Northwest National Laboratory on the drying behavior of N-Reactor spent nuclear fuel elements that had been stored underwater in the Hanford 100 Area K-Basins. The drying test series was designed to test fuel elements that ranged from intact to severely damaged.

The work discussed here was a second dry-run of the test procedure using the test apparatus without a fuel element. The purpose of this second dry-run was to establish the background levels of hydrogen in the system, and the hydrogen generation and release characteristics attributable to the test system, without a fuel element present but with water (~30 ml) added at the start of the test. This test also serves to establish the water release characteristics and background levels of water in the system during various parts of the testing. This information is used to better understand the behavior of various components of the drying system, such as particulate filters and materials used in the retort assembly, on the overall results as they apply to fuel element testing.

The dry-run test was conducted in the Whole Element Furnace Testing System located in G-Cell within the Postirradiation Testing Laboratory (PTL). The furnace testing system is composed of three basic subsystems: the in-cell furnace equipment, the system gas loop, and the analytical instrument package. This system was subjected to drying processes based on those proposed under the Integrated Process Strategy, which included a hot drying step. The test cycles are listed below:

- Cold Vacuum Drying (CVD) at ~50°C under vacuum (~2 hr)
- Pressure Rise Test at ~50°C (~1 hr)
- Hot Vacuum Drying (HVD) for a total of ~59 hr (~17 hr at ~80°C, ~33 hr at ~80°C to ~400°C, and ~10 hr at ~400°C)
- System cooldown to ~25°C (~54 hr).

Approximately 10 ml of water were observed in the condenser during the condenser pumpdown phase of CVD (of ~30 g added at the start of the test), and an additional 0.2 mg of water was removed during the post-CVD Pressure Rise Test from *free* water not completely released during CVD. Comparison of the Pressure Rise Test data with data from fuel element runs 3 and 4 suggests that other sources of gas are responsible for some of the observed total pressure rise. Most of the moisture pressure rise is attributed to water release from the furnace system.

Water removed during the three separate phases of HVD was ~79 mg, ~23 mg, and ~5 mg, respectively. An additional ~1 mg of water was released during post-HVD cooldown, indicating that the fuel

element testing system still contained small residual quantities of water after the drying test was completed. The water released after the Pressure Rise Test is attributed to the release of free water from other parts of the furnace system. As expected, the bulk of the water removal occurred during the CVD phase.

Hydrogen data were obtained from the gas chromatograph during HVD when argon was flowing through the system. As was also expected, hydrogen release rates were very low, averaging ~ 0.0005 Torr-l/min over the course of the HVD. Approximately 1.3 Torr-l (0.14 mg) of hydrogen was released over the HVD, likely from oxidation of the metal components by any remaining water or desorption of hydrogen from the metal surfaces of the furnace system.

Quality Assurance

This work was conducted under the Quality Assurance Program, Pacific Northwest National Laboratory (PNNL) SNF-70-001, *SNF Quality Assurance Program*, as implemented by the PNNL *SNF Characterization Project Operations Manual*. This QA program has been evaluated and determined to effectively implement the requirements of DOE/RW-0333P, Office of Civilian Radioactive Waste Management *Quality Assurance Requirements and Description (QARD)*. Compliance with the QARD is mandatory for projects that generate data used to support the development of a permanent High-Level Nuclear Waste repository. Further, the U.S. Department of Energy has determined that the testing activities which generated the results documented in this report shall comply with the QARD. Supporting records for the data in this report are located in the permanent PNNL SNF Characterization Project records, *Furnace Testing of Second Dry-Run*.

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Acronyms

ATS	Applied Test Systems
CVD	Cold Vacuum Drying
DACS	data acquisition and control system
ET	elapsed time
GC	gas chromatograph
HP	Hewlett Packard
HVD	Hot Vacuum Drying
ID	inside diameter
IPS	Integrated Process Strategy
MS	mass spectrometer
NIST	National Institute of Standards and Technology
OD	outside diameter
PNNL	Pacific Northwest National Laboratory
PTL	Postirradiation Testing Laboratory
QA	Quality Assurance
QARD	Quality Assurance Requirements and Description
SNF	spent nuclear fuel
UHP	ultra high purity
VP	vapor pressure

1.0 Introduction

The water-filled K-Basins in the Hanford 100 Area have been used to store N-Reactor spent nuclear fuel (SNF) since the 1970s. Because some leaks have been detected in the basins and some of the fuel is breached due to handling damage and corrosion, efforts are underway to remove the fuel elements from wet storage. An Integrated Process Strategy (IPS) has been developed to package, dry, transport, and store these metallic uranium fuel elements in an interim storage facility on the Hanford Site (WHC 1995). Information required to support the development of the drying processes, and the required safety analyses, is being obtained from characterization tests conducted on fuel elements removed from the K-Basins. A series of whole element drying tests (reported in separate documents, see Section 7.0) have been conducted by Pacific Northwest National Laboratory (PNNL)^(a) on several intact and damaged fuel elements recovered from both the K-East and K-West Basins.

This report documents the results of the second dry-run test, which was conducted without a fuel element. With the concurrence of project management, the test protocol for this run, and subsequent drying test runs, was modified. These modifications were made to allow for improved data correlation with drying procedures proposed under the IPS. Details of these modifications are discussed in Section 3.0.

This test was performed using a empty test apparatus to which approximately 30 ml of water had been added. The empty apparatus was subjected to a combination of low- and high-temperature vacuum drying treatments that were intended to mimic, wherever possible, the fuel treatment strategies of the IPS. The purpose of this second dry-run was to establish the background levels of hydrogen in the system, and the hydrogen generation and release characteristics attributable to the test system, without a fuel element present but with water added prior to the start of the test. This test also serves to establish the water release characteristics and the background levels of water in the system during the various parts of the testing cycle. These dry-run water release characteristics are used in the data interpretation for subsequent tests.

The system used for the drying test was the Whole Element Furnace Testing System, described in Section 2.0, located in the Postirradiation Testing Laboratory (PTL, 327 Building). The test conditions and methodologies are given in Section 3.0. The experimental results are provided in Section 4.0. These results are further discussed in Section 5.0.

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2.0 Whole Element Furnace Testing System

A complete description for the Whole Element Furnace Testing System, including detailed equipment specifications, is provided in Ritter et al. (1998).

2.1 Major Systems Overview

An overview of the furnace testing system is presented in this section. The subsystems pertinent to this test report are as follows:

- **Vacuum Pumping System**—This system consists of a scroll-type vacuum pump, a condenser with chiller, filters, valves, and piping, which provide the vacuum pressures and flows required for the proposed IPS vacuum processes.
- **Process Heating System**—This system consists of a resistively heated, clam-shell furnace and a sample chamber (retort) to provide heating to the fuel element and to control process temperatures.
- **Gas Supply/Distribution System**—This system consists of gas bottles; mass flow controllers; piping; and valves for metering argon, air, or oxygen through the system. A bubbler is also available for adding water vapor to the system if desired.
- **Gas Analysis Instrumentation**—The gas analysis instrumentation includes a 300-amu quadrupole mass spectrometer (MS) and a gas chromatograph (GC) for monitoring selected elements in the process gas stream.
- **Process Instrumentation**—The system is equipped with several instruments for measuring process temperatures, pressures, and moisture level. An auxiliary turbo vacuum pumping system provides low system pressures for zero adjustment of the high accuracy retort pressure sensor.
- **Data Acquisition and Control System (DACS)**—The DACS consists of an IBM-compatible computer and data acquisition/control unit to monitor/store key system parameters (temperatures, pressures, flows, moisture level), along with controlling the process heating system and a safety argon system.

Figures 2.1 and 2.2 are photographs of the equipment located inside and outside of the PTL G-Cell. The furnace (including retort), and some of the process piping, instrumentation, and valves are located inside the hot cell. The furnace sits on the cell floor, and the process piping is routed to a rack that hangs on the west cell wall. Process piping, electrical power, and instrumentation wires pass through several split plugs on the west side of the cell. The process piping on the outside of the cell is contained within a glove bag, which provides a secondary containment as a precaution in case the process piping lines become contaminated. The vacuum pump, condenser, bubbler, GC and the remainder of the

Figure 2.1. Fuel Element Drying System Components (in-cell)

Figure 2.2. Fuel Element Drying System Components (ex-cell)

instrumentation and valves are located inside this glove bag. Instrumentation and electrical power wires are routed through pass-through sleeves on the sides of the glove bag to the instrument rack and computer console.

The instrument rack contains the readout/control units for the pressure sensors, moisture sensor, and flow controllers, along with the heat trace temperature controllers, data acquisition/control unit, turbo pump controller, GC laptop computer, and uninterruptible power supplies. The computers for the DACS and MS are located next to the instrument rack. The following sections provide more detailed descriptions of the components for these subsystems.

2.2 Vacuum Pumping System

The vacuum pumping system provides the pressures and flows required for the proposed IPS processes. This system connects the furnace retort with all the other components of the test system through various valves, fittings, and piping. The vacuum pumping system consists of the following components:

- scroll pump for evacuating the system to pressures below 1 Torr
- water condenser with refrigerated chiller for gross removal of water
- valves and piping for connecting the various components and controlling the flow direction
- particulate filters to prevent the spread of contamination
- heating cords with temperature controllers for preventing condensation in lines.

2.2.1 Varian Scroll Pump

The system vacuum pump is a Varian model 300DS scroll pump. This pump has an ultimate vacuum pressure less than 10^{-2} Torr and a peak pumping speed of 250 l/min (8.8 cfm). These pressures and flows are more than adequate for simulating the conditions of the proposed IPS vacuum processes. For a single fuel element, this amount of flow may be more than desired. Therefore, a metering valve was installed on the pump inlet to throttle the flow to lower levels as required. The desired system pressure is achieved by either using the metering valve or flowing ultra high purity (UHP) argon into the system through the entire gas loop or via a direct injection of ballast gas at the pump inlet. The use of argon gas helps to prevent the in-leakage of moisture-containing air through small system leaks (which are difficult to eliminate) that would interfere with process monitoring equipment.

2.2.2 Water Condenser

The scroll vacuum pump can be damaged by condensation of liquid water in the scroll mechanism, and, since each fuel element is wet at the start of each test, the possibility of pump damage was considered. A water condenser with corresponding chiller was installed in the system to condense the bulk of

the water before it reaches the pump. This condenser can be valved into the system in series with the scroll vacuum pump or can be bypassed if not needed. The condenser cannot trap all the liberated free water, but is efficient at removing the majority of free water in the system. The condenser is only used during the first phase of a Cold Vacuum Drying (CVD) test for a single fuel element. The condenser was custom fabricated specifically for this system. Detailed sketches and specifications for the condenser are given in Ritter et al. (1998).

2.2.3 Piping, Valves, and Filters

The vacuum pumping system connects the system components through various valves, fittings, and piping. A simplified piping schematic for the system is shown in Figure 2.3. This schematic shows the basic flow path of gases through the system that was used for this test, along with the relative locations of the major components, valves, and instruments. Detailed system piping diagrams are provided in Ritter et al. (1998), along with approximate lengths for the piping lines. As seen in Figure 2.3, there are numerous valves in the system that are used to direct the flow to and from the various components. Most of the valves in the system are ball valves and range from 1/4 in. to 1/2 in. nominal size. The system piping is constructed of thin wall tubing (1/4 in. to 1/2 in. OD) and is typically connected using simple Swagelok fittings (tees, elbows, unions, etc.). Ports for gas sampling/analysis and monitoring of system pressure, temperature, and humidity are also provided at key locations in the system piping. Special fittings and pipe-threaded fittings are used in some locations for connecting piping to the process instruments.

Particulate filters are installed in the system on both the inlet and outlet to the retort to help prevent the spread of contamination to the system piping on the outside of the hot cell. These filters are constructed of a microporous fiberglass media in stainless steel housing. They are 99.9% efficient for particulates that are 0.2 microns and larger in size. Two different size filters, manufactured by Matheson, are used in the system.

2.2.4 System Line Heaters

All of the stainless steel tubing that carries gases into the furnace retort and resultant gases from the retort is heated to about 75°C to ensure condensable water vapor remains in the gas phase. Simple heat "cords" capable of being wrapped upon each other (as required at tees, elbows, and other connections) were found to be a good heating method for this system. Simple proportional controllers control the heating cords. Type-K thermocouples are installed on each heated line so the DACS can be used to monitor and record temperature.

2.3 Process Heating System

The whole element furnace is a 4-ft-long, resistively heated, clam-shell furnace. The furnace, Series 3210 supplied by Applied Test Systems (ATS), has a temperature rating of 900°C and total heating capacity of 13,800 W. The internal dimensions are 5 in. ID by 45 in. long. The furnace has three separate sets of heating elements that allow the heating to be controlled in zones; each zone is 15 in. long and supplies up to 4600 W heating. The zones can be controlled separately to establish a flat temperature profile within the furnace, even though heat is lost preferentially out the end with the retort entry flange. A heat

Figure 2.3. Generalized View of Test System

reflector consisting of several thin Inconel plates is used to reduce heat loss from the flange end of the retort. The furnace controller is an ATS Series 3000, which consists of three programmable, self-tuning proportional with integral and derivative controllers. These controllers are also interfaced to the DACS, which is capable of providing limited input to the controllers as required. The retort, an ATS Series 3910, is an Inconel tube fitted with a gas inlet tube at one end and a gasketed flange at the other. Of all high-temperature materials, Inconel series 600 was selected to reduce the amount of oxidation and water pickup by the retort and associated components. Experience has shown that stainless steel components were easily affected by corrosion, which could then affect test results. The body of the retort is fabricated from schedule 40 Inconel pipe (4.5 in. OD, 4.026 in. ID), and the inside length is about 44.5 in. Seven type-K thermocouples are installed equidistant along one side of the retort and extend into the retort interior approximately 1/8 in. These thermocouples are used to monitor the retort temperature so that if a reaction with the fuel element occurs (which would locally raise the retort temperature), this event can be correlated with the approximate location on the fuel.

An Inconel sample/transfer boat is used to load the fuel element into the furnace. The boat is fabricated from an 11-gauge (0.120-in.-thick) Inconel 601 sheet, which is formed into a flattened u-shape. The boat has a weir and a swivel handle on each end. The weirs are used to keep free water or particulates contained in the boat as required.

2.4 Gas Supply/Distribution System

The gas supply system and vacuum pumping system together are capable of controlling the fuel element environment to vacuum or moderate pressure conditions, and/or exposing the fuel element to a variety of gases or gas mixtures. The gas loop is typically operated as a single-pass system with no capability for recirculation. The gas supply system consists of gas bottles; mass flow controllers; piping; and valves for metering argon, air, or oxygen through the system. A bubbler is also available for adding water vapor to the process gas stream as required, but it was not used in these tests.

The gas supply system contains three Matheson mass flow controllers calibrated for argon, air, and oxygen. All gases are typically specified "ultra high purity" and are additionally filtered for water using molecular sieve columns. Argon is the principal inert gas used, as it is denser than air; provides reasonable thermal conductivity; and requires simpler handling procedures than lighter gases such as helium. The argon purge gas is introduced into the retort through FE-01, which is a Matheson model 8272-0422 oxygen controller, recalibrated for argon gas at 25°C using a NIST-traceable bubble flow meter. The recalibration resulted in a flow rate range of 0 - 324 sccm argon. Air and oxygen are not currently used because any oxidative steps have been deleted from the current IPS for the SNF. The manufacturer's specifications for the air and oxygen controllers' flow rate ranges are 0 - 1000 sccm air and 0 - 10 sccm oxygen. If higher flow rates are desired, a new mass flow controller with a higher range could be procured and installed in the system.

2.5 Gas Analysis Instrumentation

2.5.1 Balzers Omnistar Mass Spectrometer

The Balzers Omnistar Mass Spectrometer is a compact, computer-controlled, quadrupole MS capable of scanning to 300 amu. The unit is capable of monitoring up to 64 components within a gas stream with a nominal detection limit of less than 1 ppm for most gases other than hydrogen. The MS was used to monitor hydrogen, nitrogen (for air in-leakage), krypton, xenon, and other elements during the test.

The MS was modified as a result of early system testing and calibration to improve the time response to small changes in hydrogen pressure. Prior to testing, the MS was calibrated for hydrogen using mixtures of hydrogen and helium, and hydrogen and argon gas. The residence time of each gas could be measured in the quadrupole chamber, and it was observed that the hydrogen decay time was approximately four times as long as helium. This was not unexpected as turbomolecular pumps have a lower pumping efficiency for very light gases. In standard practice this is acceptable, but for these tests, where determining hydrogen could be very important, steps were taken to improve the hydrogen decay time. The MS vacuum system was modified by adding a stainless steel flanged tee, a gate valve, and a room-temperature hydrogen getter downstream from the quadrupole. Under vacuum the gate valve can be opened, exposing the getter to the system to help scavenge hydrogen from the system following analysis. This modification reduced the residence time of hydrogen in the system substantially, and decreased the background level of hydrogen by about a factor of 2. The getter improved the system response to transient events that might result in the release of hydrogen.

A Granville-Phillips variable leak valve, series 203, was added to the gas sampling inlet of the MS in order to permit operation over a wide range of system pressures. Without the leak valve, system pressures above about 40 Torr produce too much flow through the MS capillary tube, which overwhelms the turbo pump used to pump down the MS vacuum chamber. Flow through the leak valve can be continuously varied from 0.4 l/s to 10^{-11} l/s, which allows the MS inlet pressure to be controlled to any pressure desired, even if the system pressure varies dramatically. The pressure on the low pressure side of the leak valve was measured using a Cole-Parmer sensor (PE-07) and recorded by the DACS. The pressure used for the calibration divides the inlet head pressure, and this factor is applied to the test data for calculating actual gas concentrations. The MS was calibrated at ~30 Torr head pressure with a certified gas standard consisting of 1050 ppmv hydrogen in argon.

2.5.2 MTI M200 Gas Chromatograph

The MTI M200 Gas Chromatograph is a high-speed GC that is used to monitor the quantities of hydrogen and other light gases in the furnace testing system gas loop. This instrument is interfaced with a laptop computer to record data. The GC is designed to operate at near-atmospheric pressure; thus it may be configured in two different ways for measurement purposes. At system pressures near atmospheric, the GC is configured to sample directly from the gas loop ahead of the system vacuum pump (as was done in the first two fuel element tests, but not in this dry-run protocol). When the system is under vacuum, the GC is configured to sample from the exhaust side of the vacuum pump. The gas output from the pump is sufficiently compressed that the GC can sample and analyze this gas. The GC inlet

pressure is measured using a Cole-Parmer pressure sensor (PE-08) and recorded by the DACS. No correction for the difference in the sample pressure and calibration pressure is applied, since both are ~760 Torr (1 atm). The GC was calibrated with a certified gas standard consisting of 114 ppmv hydrogen in synthetic air.

2.6 Process Instrumentation

The furnace testing system contains several process instruments for monitoring moisture content, pressure, and temperature. The key instruments are as follows:

- Panametrics moisture monitor
- MKS Baratron pressure transducers
- Cole-Parmer pressure transducers
- Type-K thermocouples.

2.6.1 Panametrics Moisture Monitor

The Panametrics moisture monitor model MMS35 uses a solid electrochemical probe (model M2L) that measures moisture by measuring the characteristic capacitance of the probe as a function of the moisture in the gas phase. The sensor has a nominal dew point range of -110°C to 20°C. Previous testing indicated that contamination of the probe by radioactive elements (e.g., cesium) causes the probe to lose calibration and results in moisture readings that drift with time. To prevent contamination of the probe tip, the probe is installed in the gas loop downstream of two glass particulate filters. Further, the probes are changed following each test and surveyed for radioactive contamination. If no contamination is found, and the data correlate well with the data obtained from the MS, the readings are accepted.

A calibration verification procedure can be performed using calibrated water "leak" tubes. These tubes can be placed inside the furnace and, when heated, will establish a known water vapor pressure in the system. However, this procedure is time intensive; approximately 2 weeks are required to calibrate one probe over the range of moisture likely to be encountered in these tests. This procedure is only used if the moisture monitor results vary widely from the MS data.

Output of the moisture monitor is in dew point in degrees Celsius. For comparison with other test data, these dew point values were converted to water vapor pressure in Torr using the water and ice vapor pressure data shown in Table 2.1. Interpolation of the data was accomplished using a 6th-order polynomial fit to the log of the vapor pressure (VP) versus temperature data. The resulting conversion expression is as follows:

Table 2.1. Water and Ice Vapor Pressure Data Versus Temperature

Dew Point (°C)	Vapor Pressure (VP)		
	(Pa) ^(a)	(Torr)	Log (Torr)
-80	5.5000E-02	4.136E-04	-3.3834
-75	1.2200E-01	9.174E-04	-3.0374
-70	2.6100E-01	1.963E-03	-2.7071
-65	5.4000E-01	4.061E-03	-2.3914
-60	1.0800E+00	8.122E-03	-2.0904
-55	2.0930E+00	1.574E-02	-1.8030
-50	3.9360E+00	2.960E-02	-1.5287
-45	7.2020E+00	5.416E-02	-1.2663
-40	1.2840E+01	9.656E-02	-1.0152
-35	2.2350E+01	1.681E-01	-0.7745
-30	3.8010E+01	2.858E-01	-0.5439
-25	6.3290E+01	4.759E-01	-0.3224
-20	1.0326E+02	7.765E-01	-0.1099
-15	1.6530E+02	1.243E+00	0.0945
-10	2.5990E+02	1.954E+00	0.2910
-5	4.0176E+02	3.021E+00	0.4802
0	6.1129E+02	4.597E+00	0.6625
10	1.2281E+03	9.235E+00	0.9655
(a) CRC Press. 1997. <i>Handbook of Chemistry & Physics</i> , 78 th edition.			

$$VP(\text{Torr}) = \log^{-1}(Ax^6 + Bx^5 + Cx^4 + Dx^3 + Ex^2 + Fx + G) \quad (2.1)$$

where x = the measured dew point (°C) and

$$A = -6.7260 \cdot 10^{-12}$$

$$B = -1.7250 \cdot 10^{-9}$$

$$C = -1.7089 \cdot 10^{-7}$$

$$D = -7.2618 \cdot 10^{-6}$$

$$E = -2.9668 \cdot 10^{-4}$$

$$F = 3.4414 \cdot 10^{-2}$$

$$G = 0.66043$$

2.6.2 MKS Baratron Pressure Transducers

Two MKS Baratron model 690 calibrated pressure transducers coupled with MKS model 270 signal conditioners are used as the primary measurement for the overall system pressure. As shown in Figure 2.3, PE-01 measures the system pressure downstream of the retort outlet, whereas PE-06 measures the system pressure at the retort inlet. PE-01 indicates pressure in the range of 0.1 Torr to 10,000 Torr. The pressure range of PE-06 is 0.01 Torr to 1000 Torr. PE-06 was installed after the first two fuel

element drying tests to provide more accurate measurements than PE-01 for low pressures. PE-06 is therefore considered the primary system pressure measurement. In addition, the 270 signal conditioner procured with PE-06 has a special capability to remotely zero the transducer, which provides more accurate pressure measurements below 1 Torr.

An auxiliary high vacuum turbo pump is used to evacuate the inlet to PE-06 to well below 10^{-4} Torr, so that the transducer can be accurately re-zeroed. The 270 signal conditioner used with PE-01 does not have a remote zeroing capability. Both signal conditioners have analog outputs that are interfaced to the DACS so that system pressure is continuously recorded.

2.6.3 Cole-Parmer Pressure Transducers

Two Cole-Parmer model H-68801-53 diaphragm-type, calibrated pressure transducers are installed on the MS and GC sample lines, as indicated by PE-07 and PE-08 in Figure 2.3. These pressure measurements are used to normalize the MS and GC data so that actual gas concentrations in the system can be calculated from the relative concentrations measured. These sensors have a range of 0 to 1500 Torr with a resolution of 0.1 Torr, and an accuracy of $\pm 1\%$ or ± 1 Torr, whichever is larger. Both readout units (model H-68801-03) have analog outputs that are interfaced to the DACS so that these pressures are continuously recorded.

2.6.4 Thermocouples

Thermocouples provide a simple, reliable method for measuring system temperatures. As shown in Figure 2.3, over 20 thermocouples are installed at various locations in the system to provide key temperature measurements. The retort temperatures are of primary importance, and these temperatures are measured by thermocouples TE-04 through TE-10, which are positioned equidistant along the length of the retort. Other key temperature measurements include the retort center temperature (TE-20, which is a 30-in.-long thermocouple installed through the outlet end of the retort), retort inlet temperature (TE-21), condenser gas temperature (TE-19), and the condenser coolant temperature (TE-22). Thermocouples TE-11 through TE-17 are used for controlling the temperature of the heated lines. All thermocouple readings are continuously recorded using the DACS.

2.7 Data Acquisition and Control System

The DACS monitors system parameters, and controls the furnace and the safety argon system. The DACS consists of a Hewlett Packard (HP) 3497A data acquisition/control unit, and an IBM-compatible computer. A National Instruments general purpose interface bus card, installed in the IBM-compatible computer, is used to communicate with the HP 3497A. The computer communicates with the furnace temperature controllers over serial port 0 using an RS-232/RS-485 converter. The DACS uses National Instruments LabView for Windows as the control software.

The DACS is designed to measure critical system parameters during fuel conditioning tests, including temperatures, pressures, flow rates, and moisture level. The measured parameters are converted to engineering units, displayed on the computer screen, and stored to disk at user-defined intervals. The data

files are stored in a tab-delimited format to allow importing into a standard spreadsheet or plotting program. A plotting screen also allows for plotting of up to six parameters at a time.

Limited control of the furnace can be performed with the DACS. Each of the three furnace zone temperatures can be remotely set by the DACS. In addition, the DACS allows the operator to start and stop the furnace and select one of four temperature profiles that are pre-programmed in the furnace temperature controllers. Note that these profiles must be programmed manually in the furnace controllers before using the DACS to set them.

3.0 Vacuum Drying Testing of the Furnace System

The test was performed in accordance with the requirements of *Test Plan for Whole Element Furnace Run 4*, SNFCT98:012:R00, and the individual Test Procedure, *Dry Run Testing of G-Cell Furnace Using a Water Sample*, PTL-003, Revision 0. These documents are located in the PNNL permanent project records for this test.

The testing consisted of two parts (discussed in this section):

- Drying the system using a combination of Cold Vacuum Drying (CVD) and Hot Vacuum Drying (HVD) processes.
- Cooling the furnace, and returning the system to a state ready for whole fuel element testing.

3.1 Initial Conditions

Thirty milliliters of water were added to the sample boat. The test conditions used were essentially the same as those used for fuel elements 0309M, 5744U, 6603M, 1164M, 2660M, and 6513U (Runs 3 through 8, see Section 7.0).

3.2 System Drying

The test protocol for the second dry-run was changed from that used in the first dry-run. These modifications were made to allow for improved data correlation with drying procedures proposed under the IPS. These changes largely involved the argon flow and vacuum pump conditions that existed during various phases of the test. For example, the CVD step was now run without argon flow, and the temperature rise phase of HVD was now run with vacuum pumping. Earlier tests had been run using a low argon flow rate during CVD and no vacuum pumping during the temperature rise phase of HVD. Other small modifications to the protocol were also made.

For the new protocol, the empty furnace system (and fuel elements during subsequent runs) was subjected to both hot and cold vacuum drying steps conducted in six phases:

1. Cold Vacuum Drying
2. Pressure Rise Test
3. Hot Vacuum Drying (first step)
4. Hot Vacuum Drying (second step)
5. Hot Vacuum Drying (third step)
6. Cooldown.

The conditions used for these test phases are summarized in Table 3.1. Each phase is described below.

Table 3.1. Summary of Nominal Test Design Conditions

Test Segment	Nominal Test Condition ^(a)
A. Cold Vacuum Drying System Configuration Test Temperature, °C Atmosphere Pressure, Torr Gas Flow Rate, cc/min Gas Species Monitored Duration, hr	Pump on, no argon flow 50 Vacuum <5 0 H ₂ , H ₂ O, N ₂ , O ₂ , CO ₂ , Ar, Kr, Xe CVD will be conducted until the total pressure in the retort falls below 0.5 Torr.
B. Pressure Rise Test System Configuration Test Temperature, °C Atmosphere Initial Pressure, Torr Gas Flow Rate, cc/min Gas Species Monitored Pressure Rise (acceptable levels, Torr) Duration, hr	Test Chamber Isolated 50 Vacuum <5 0 H ₂ , H ₂ O, N ₂ , O ₂ , CO ₂ , Ar, Kr, Xe <0.5 1
C. Hot Vacuum Drying (Step 1) System Configuration Test Temperature Range, °C Atmosphere Pressure, Torr Gas Flow Rate, cc/min Gas Species Monitored Duration, hr	Pump on, argon gas flow 75 Vacuum, Ar background 15 300 H ₂ , H ₂ O, N ₂ , O ₂ , CO ₂ , Ar, Kr, Xe 24
D. Hot Vacuum Drying (Step 2, heatup) System Configuration Test Temperature Range, °C Temperature Ramp Rate, °C/hr Atmosphere Pressure, Torr Gas Flow Rate, cc/min Gas Species Monitored Duration	Pump on, argon gas flow 75 to 400 10 Vacuum, Ar background 15 300 H ₂ , H ₂ O, N ₂ , O ₂ , CO ₂ , Ar, Kr, Xe 35

Table 3.1. (contd)

Test Segment	Nominal Test Conditions ^(a)
E. Hot Vacuum Drying (Step 3)	
System Configuration	Pump on, argon gas flow
Test Temperature, °C	400
Atmosphere	Vacuum, Ar background
Pressure, Torr	15
Gas Flow Rate, cc/min	300
Gas Species Monitored	H ₂ , H ₂ O, N ₂ , O ₂ , CO ₂ , Ar, Kr, Xe
Duration, hr	10
F. Cooldown	
System Configuration	Pump on, argon gas flow
Test Temperature, °C	400 to 50
Atmosphere	Vacuum
Initial Pressure, Torr	15
Gas Flow Rate, cc/min	300
Gas Species Monitored	H ₂ , H ₂ O, N ₂ , O ₂ , CO ₂ , Ar, Kr, Xe
Duration, hour	~100
(a) Nominal test design conditions. Actual values are given in the text.	

3.2.1 Cold Vacuum Drying

Air was first introduced into the furnace by allowing the ambient hot cell air to flow into the retort, as would occur during the loading of a fuel element. The furnace temperature was then increased to approximately 50°C and allowed to stabilize. The system vacuum pump was then turned on and the system water condenser valved in. When the system pressure dropped to a reasonable level, the vacuum pump was valved out and only the condenser left in the system. When the system pressure reached the vapor pressure of water, the condenser was valved out of the gas loop, and the vacuum pump was used to remove any free water from the system. During this time, the pressure dropped steadily until the water vapor pressure became lower than the condenser could extract. At this point, the water content in the gas phase began to rise, and the condenser was valved out of the system while the vacuum pump was valved back in to continue evacuating the system. No argon was injected during CVD in this configuration. CVD was conducted for ~17 hr at an ultimate pressure of ~0.4 Torr. The purpose of this phase of the test was to determine if CVD is successful in removing the majority of the free water from the system in a reasonable length of time.

3.2.2 Pressure Rise Test

The Pressure Rise Test involved isolating the system from the effects of vacuum pumping and measuring any system pressure increases while at CVD pressure and temperature conditions. The purpose of the Pressure Rise Test was to determine the effectiveness of the preceding CVD process. This test was

conducted by valving the vacuum pump out of the gas loop and closing the exhaust valves in the gas loop. The condition for acceptance of this portion of the test is a total system pressure rise of less than 0.5 Torr in a 1-hr time period. If this condition is not met, the system is re-opened to the vacuum pump and the Pressure Rise Test repeated.

3.2.3 Hot Vacuum Drying, Step 1

Following completion of the Pressure Rise Test, the vacuum pump was re-opened to the system retort; argon gas flow was established at a rate of 324 cc/min; and the retort temperature was increased to $\sim 80^{\circ}\text{C}$. This condition was held for a period of ~ 17 hr. This portion of the test can be used to obtain isothermal hydrogen and water release data for assessing oxidation of the fuel at low temperatures.

3.2.4 Hot Vacuum Drying, Step 2

The second step of the HVD test involved raising the temperature of the retort from $\sim 80^{\circ}\text{C}$ to $\sim 400^{\circ}\text{C}$ at a carefully controlled rate while maintaining the same argon flow and pressure conditions. Thus, any release of gas species during this temperature rise can be assigned to a specific temperature. The second step of the HVD process was conducted for about 33 hr.

During actual fuel element drying, hydrogen may be released from a fuel element through the decomposition of uranium hydride. Water may also be liberated by various hydrated species found on fuel elements, such as hydrates of uranium oxides, aluminum hydroxides, and hydrated iron oxides, and also released slowly from beneath corroded parts of the fuel element and from behind the cladding. The released water can also react with the fuel element to generate hydrogen.

3.2.5 Hot Vacuum Drying, Step 3

The final step of the HVD process involved holding the temperature of the retort at $\sim 400^{\circ}\text{C}$ while again maintaining the same argon flow and pressure conditions as in steps 1 and 2. The hold point will yield isothermal release data for any remaining hydrated species on actual fuel elements, and for oxidation of uranium by remaining water. This step of the HVD process was conducted for about 10 hr.

3.2.6 System Cooldown and Second Pressure Rise Test

Following completion of the final HVD step, the system retort was allowed to cool to $\sim 50^{\circ}\text{C}$. Data collection was continued through the cooldown phase.

3.3 Calculation of Water and Hydrogen Inventories

Assuming ideal gas behavior of the water vapor, the total water inventory (in grams) in the system during those portions of the test conducted with argon flowing into the retort can be approximated from the measured water vapor pressure and the argon gas flow as follows:

$$\frac{dm}{dt} = \frac{M}{V_0} \cdot \frac{P_w}{(P_t - P_w)} \cdot \frac{dV}{dt} \quad (3.1)$$

where dm/dt is the rate of water removal in grams per minute, M is the molecular mass of water in grams per mole, dV/dt is the flow rate in liters per minute (at the calibration temperature of 25°C), V_0 is the molar volume of gas at 25°C and 1 atmosphere in liters per mole, P_w is the partial pressure of water vapor in Torr, and P_t is the total pressure in Torr. The total amount of water released is given by integrating the rate data over time.

The hydrogen inventory may be calculated in a similar fashion with the $[P_w/(P_t - P_w)]$ expression in the above equation replaced with the measured atom fraction of hydrogen. For the purposes of this report, all hydrogen data are plotted in Torr·l rather than grams. At the calibration condition of the argon flow controller, 1 Torr·l is equivalent to approximately 0.11 mg of hydrogen.

The assumptions made in estimating the water and hydrogen values are:

- The flow into the retort is approximately equal to the flow out (i.e., contributions to the flow from other gas species such as hydrogen are neglected).
- The argon mass flow is referenced to 25°C (as determined from the calibration of the flow gauges)
- The sample gas is at the same temperature as the calibration gas (GC and MS measurements).

4.0 Experimental Results

In the following sections, the experimental data collected during the drying test are expanded and plotted for each segment. Summary results from the test are plotted in Figure 4.1. This figure shows the system moisture level response to the pressure changes and the retort tube temperatures during the test. Time intervals for the various test segments are shown in the upper section of the plot, and also outlined in Table 4.1. The temperatures shown in Figure 4.1 were recorded from one of seven thermocouples (TE-07) on the system located near the center of the retort. The pressure data were taken from the 0 to 1000 Torr Baratron sensor (PE-06) located upstream of the retort.

4.1 Cold Vacuum Drying

The water release from the CVD portion of the dry-run is shown in Figure 4.2. The baseline moisture partial pressure in the system prior to heating was ~7 Torr at a retort temperature of ~28°C. Total system pressure was ~744 Torr, with no argon gas flow. After the system was heated to ~50°C, the moisture pressure remained at ~7 Torr and the system pressure rose to ~827 Torr. Based on the pressure differences at these two temperatures and assuming ideal gas behavior, there is an approximately 29 Torr pressure excess. This excess pressure may be due to gases evolved during the heatup to 50°C, such as hydrogen, and gases dissolved in the free water. Another explanation for the calculated pressure difference is that the average retort temperature was somewhat greater than 50°C.

The CVD commenced at an elapsed time (ET) of 194 min when the vacuum pump was opened briefly in order to "rough-out" the system retort to ~17 Torr (total pressure). Figure 4.2 shows that the moisture pressure initially rose, reaching a maximum of about 11 Torr after several minutes, then dropped slowly to ~4 Torr over the next 80 min while the condenser and vacuum pump were being used alternately to remove most of the water. At ~275 min ET, the condenser was valved out of the system for the final time, and the retort opened to the vacuum pump. At this point, both the moisture pressure and the total pressure dropped more rapidly, with both pressures reaching a final value of ~0.4 Torr by the end of CVD. The temperature of the system dropped from ~53°C to about 48°C during the pumpdown when most of the moisture was removed. Approximately 10 ml of water were observed in the condenser during CVD. Because of the design of the condenser, however, the observed water is likely to be an under-estimation of the total water collected.

4.2 Pressure Rise Test

The results of the pressure rise phase of the drying test are shown in Figure 4.3. As discussed above, the data plotted for the total pressure are from the 0 to 1000 Torr Baratron sensor (PE-06) located upstream of the retort. This sensor has higher sensitivity and therefore lower noise than the 0 to 10,000 Torr Baratron (PE-01) located downstream of the retort. For calculation of total water mass removed from the retort, pressure data from the 0 to 10,000 Torr sensor were used, as this sensor is closer

Figure 4.1. Second Dry-Run, Summary Plot

Table 4.1. Second Dry-Run Time Line

Activity	Date/Time	Elapsed Time (min)
Start of Test		
Heat furnace to ~50°C	11/26/97 10:42	37
Cold Vacuum Drying Test		
Open pump valve and condenser to furnace (initial)	11/26/97 13:19	194
Open pump valve, close condenser valve (final)	11/26/97 14:40	275
Pressure Rise Test(s)		
Close pump (isolate furnace)	11/26/97 14:54	289
Open pump valve	11/26/97 15:56	351
Hot Vacuum Drying Test (Step 1)		
Start argon flow (~320 cc/min), raise furnace temperature to ~80°C and hold	11/26/97 15:57	352
Hot Vacuum Drying Test (Step 2)		
Raise furnace temperature to ~400°C @ 10°C/min	11/27/97 08:39	1354
Hot Vacuum Drying Test (Step 3)		
Hold furnace temperature at ~400°C	11/27/97 17:12	3308
System Cooldown		
Reduce temperature of retort to ~50°C, maintain argon flow	11/29/97 03:03	3900
End test	12/01/97 08:54	7129

to the moisture sensor. Differences in the pressure indications of the two sensor are small, however, and therefore negligible when compared with overall system uncertainties.

The Pressure Rise Test was conducted over an ET of 289 min to 351 min. The total pressure showed an essentially linear pressure rise over the course of the test. A regression fit (dotted line in the figure) yielded a total pressure rise rate of ~0.29 Torr/hr (well below the 0.5 Torr/hr criterion for the test). The moisture pressure showed an initial non-linear increase followed by a longer linear region. The slope of the linear region was ~0.022 Torr/hr. The irregularities seen in the total pressure data are due to under-sampling of the data output from the Baratron sensor. The data acquisition software has been modified for future tests to avoid this situation.

Assuming that the water vapor pressure increase is from water sources within the test system, and assuming ideal gas behavior of the water vapor, the rate of desorption of the water will be constant, given by:

$$\frac{dn}{dt} = \frac{V}{RT} \cdot \frac{dP}{dt} \quad (4.1)$$

Figure 4.2. Second Dry-Run, Cold Vacuum Drying

where n is the number of moles of gas, V is the volume of the system ($\sim 10,000 \text{ cm}^3$), R is the gas constant ($82.06 \text{ cm}^3 \cdot \text{atm/g-mol} \cdot \text{K}$), T is the temperature ($\sim 326 \text{ K}$), and dP/dt is the rate of change in the pressure given by the slope of the regression line. The total amount of water released to the system during the Pressure Rise Test is given by the integral of the above equation. Assuming a total period of 62 min, the total amount of water released was $\sim 0.2 \text{ mg}$. Using a total surface area of $\sim 6000 \text{ cm}^2$ for the system (total surface area of the retort, sample boat, and tubing), and 10^{15} atoms per cm^2 as the monolayer gas density on surfaces, approximately one monolayer equivalent of H_2O was evaporated.

The ratio of the water pressure rise to the total pressure rise is ~ 0.08 , which is significantly higher than would be expected just from humidity alone in air in-leakage from the cell environment (air at 20°C and 25% relative humidity would yield a water pressure to total pressure ratio of ~ 0.007). This suggests that most of the observed moisture increase is from *free* water release from the empty sample boat. The observed total pressure rise rate of $\sim 0.29 \text{ Torr/hr}$ is also higher than a value of $\sim 0.11 \text{ Torr/hr}$ observed in fuel element runs 3 and 4. This suggests that other sources of gas are responsible for some of the observed total pressure rise in the Pressure Rise Test. Air in-leakage is a likely cause of some of the overall pressure rise. Subsequent tests included a 50°C post-run Pressure Rise Test in order to estimate the air in-leakage rate.

4.3 Hot Vacuum Drying

The first segment of HVD, shown in Figure 4.4, includes the ramp and hold from $\sim 50^\circ\text{C}$ to $\sim 80^\circ\text{C}$ in flowing argon gas (324 cc/min) under partial vacuum. HVD-1 occurred over an ET of 352 min to 1354 min. The moisture pressure decreased steadily from $\sim 0.4 \text{ Torr}$ to $\sim 0.6 \text{ mTorr}$ during the $\sim 80^\circ\text{C}$ period. Total system pressure was essentially constant over this first HVD phase at $\sim 19 \text{ Torr}$. Total water removed was $\sim 79 \text{ mg}$.

The second phase of HVD involved maintaining the same system conditions as in HVD-1 but raising the temperature from $\sim 80^\circ\text{C}$ to $\sim 400^\circ\text{C}$ at a rate of 10°C/hr . HVD-2 occurred over an ET of 1354 min to 3308 min. Total system pressure remained nearly the same at $\sim 19 \text{ Torr}$. As is shown in Figure 4.5, the moisture pressure also remained constant at $\sim 0.6 \text{ mTorr}$, with a small periodic variation of about 0.1 mTorr . These "peaks," occurring at $\sim 170^\circ\text{C}$ and $\sim 380^\circ\text{C}$, may be due to release of water from some hydrated metal oxides; however, it is difficult to identify a likely source since the peaks are just discernible above the background moisture levels. Total water removed during the second phase of HVD was $\sim 23 \text{ mg}$, approximately one third of that removed during the first phase.

The final phase of HVD is shown in the left-hand side of Figure 4.6 (ET of 3308 min to 3900 min), and covered the temperature hold period at $\sim 400^\circ\text{C}$. This period is characterized by a steady decrease in the moisture pressure from $\sim 0.6 \text{ mTorr}$ to $\sim 0.3 \text{ mTorr}$. Total water removed was $\sim 5 \text{ mg}$.

Following the final HVD phase, the system heaters were turned off and the system allowed to cool. Data collection continued during this cooldown period, as shown in the right-hand side of Figure 4.7. As is evident, there is a sudden drop in both the moisture and total pressure data at an ET of $\sim 4197 \text{ min}$. The

Figure 4.3. Second Dry-Run, Post-CVD Pressure Rise Test

Figure 4.4. Second Dry-Run, Hot Vacuum Drying – Step 1

Figure 4.5. Second Dry-Run, Hot Vacuum Drying – Step 2

Figure 4.6. Second Dry-Run, Hot Vacuum Drying – Step 3

Figure 4.7. Second Dry-Run, Hydrogen Release During HVD and Cooldown

reason for this drop is not clear, but it could be related to a sudden sealing of an air leak into the system. The sharp drop in the system moisture occurs at $\sim 250^{\circ}\text{C}$. Water removed during the system cooldown was ~ 1 mg.

4.4 Gas Chromatograph Measurements

The GC is calibrated to measure hydrogen in the sample gas. As discussed earlier, the hydrogen concentration is converted from ppmv to Torr-l so that the absolute quantity of hydrogen gas released can be determined independent of argon flow rate. The GC yielded data for the release of hydrogen in the gas stream during the HVD steps. During these parts of the test, argon was flowing through the system at a rate of about 324 cc/min. It was not possible to sample for hydrogen using the GC during the CVD portion of the test since it was conducted with no flow of argon gas.

Measured hydrogen release during the HVD segments of the drying test are shown in Figure 4.7. Hydrogen release rates are very low, varying from ~ 0.0003 Torr-l/min to ~ 0.0006 Torr-l/min over the course of the HVD phase. Hydrogen release generally followed the temperature profile, with a small increase starting at a retort temperature of $\sim 350^{\circ}\text{C}$, followed by a leveling off at 400°C . Approximately 1.3 Torr-l (0.14 mg) of hydrogen was released over a time period of ~ 59 hr.

The negative "spikes" in the hydrogen data are regarded as "noise transients" and thus are not interpreted as hydrogen "gettering" events.

4.5 Mass Spectrometer Measurements

The Balzers Omnistar MS was used to collect hydrogen and other gas release data over the test run. Although not shown in Figure 4.7, the MS hydrogen signal showed the same general trends as the GC data.

5.0 Discussion

Approximately 10 ml of water were observed in the condenser during the condenser pumpdown phase of CVD (~30 ml were added at the start of the test), with an additional ~0.2 mg of water being removed during the post-CVD Pressure Rise Test. This latter release can likely be interpreted as coming from *free* water that was not completely released during CVD. Comparison of the total pressure rise from the post-CVD Pressure Rise Test with that observed for fuel element runs 3 and 4 suggests that other sources of gas are responsible for some of the observed total pressure rise.

During the first segment of HVD, approximately ~79 mg of water were removed at temperatures below 80°C. The second phase of HVD released approximately 23 mg of water. The final phase of HVD at 400°C released only about 5 mg of water, with an additional ~1 mg of water released during post-HVD cooldown. This indicates that the fuel element testing system contained small residual quantities of water even after the drying test was completed. The water released after the post-CVD Pressure Rise Test is attributed chiefly to the release of free water from other parts of the furnace system. The bulk of the water removal occurred during the CVD phase.

Hydrogen data were obtained from the GC during HVD when argon was flowing through the system. Hydrogen release rates are very low, averaging ~0.0005 Torr·l/min over the course of the HVD phase. Hydrogen release generally followed the temperature profile, with a small increase starting at a retort temperature of ~350°C, followed by a leveling off at 400°C. Approximately 1.3 Torr·l (0.14 mg) of hydrogen was released over the HVD phase, likely from oxidation of metals by water, and possibly from desorption from some of the metal surfaces of the furnace system. Hydrogen data collected by the MS system showed the same general trends as the GC data. The hydrogen released by the empty test system is several orders of magnitude less than that observed during testing to date using fuel elements.

The difference between the pressure rise slope in the total pressure and the water vapor partial pressure during the Pressure Rise Test and the sudden change in the moisture level and total pressure during cooldown may have been due to air in-leakage. A post-run Pressure Rise Test was incorporated into the protocol for subsequent tests in order to assess in-leakage at 50°C.

6.0 References

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7.0 Supporting Documents and Related Reports

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Reports are written separately for the whole element drying test series as follows:

System Design Description for the Whole Element Furnace Testing System

Spent Fuel Drying System Test Results (First Dry-Run)

Spent Fuel Drying System Test Results (Second Dry-Run)

Spent Fuel Drying System Test Results (Third Dry-Run)

Drying Results of K-Basin Fuel Element 1990 (Run 1)

Drying Results of K-Basin Fuel Element 3128W (Run 2)

Drying Results of K-Basin Fuel Element 0309M (Run 3)

Drying Results of K-Basin Fuel Element 5744U (Run 4)

Drying Results of K-Basin Fuel Element 6603M (Run 5)

Drying Results of K-Basin Fuel Element 1164M (Run 6)

Drying Results of K-Basin Fuel Element 2660M (Run 7)

Drying Results of K-Basin Fuel Element 6513U (Run 8)