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Interim Results from UO_2 Fuel Oxidation Tests in Air

August 1987

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Pacific Northwest Laboratory
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INTERIM RESULTS FROM UO_2 FUEL
OXIDATION TESTS IN AIR

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ABSTRACT

An experimental program is being conducted at Pacific Northwest Laboratory (PNL) to extend the characterization of spent fuel oxidation in air. To characterize oxidation behavior of irradiated UO_2 , fuel oxidation tests were performed on declad light-water reactor spent fuel and nonirradiated UO_2 pellets in the temperature range of 135 to 250°C. These tests were designed to determine the important independent variables that might affect spent fuel oxidation behavior. The data from this program, when combined with the test results from other programs, will be used to develop recommended spent fuel dry-storage temperature limits in air. This report describes interim test results.

The initial PNL investigations of nonirradiated and spent fuels identified the important testing variables as temperature, fuel burnup, radiolysis of the air, fuel microstructure, and moisture in the air. Based on these initial results, a more extensive statistically designed test matrix was developed to study the effects of temperature, burnup, and moisture on the oxidation behavior of spent fuel. Oxidation tests were initiated using both boiling-water reactor and pressurized-water reactor fuels from several different reactors with burnups from 8 to 34 GWd/MTU. A 10^5 R/h gamma field was applied to the test ovens to simulate dry storage cask conditions. Nonirradiated fuel was included as a control. This report describes experimental results from the initial tests on both the spent and nonirradiated fuels and results to date on the tests in a 10^5 R/h gamma field.

SUMMARY

Oxidation tests with light-water reactor (LWR) spent fuel were conducted to develop a data base for design and operating conditions of facilities for receiving, handling, packaging, and storing spent LWR fuel in an air atmosphere; to predict when cover-gas purity monitoring of inert gas storage facilities is no longer required; and to provide analytical results for determining the effect of abnormal events on the fuel. The results from the initial scoping investigations led to the selection of a statistically designed spent fuel test matrix with the following variables: temperature levels from 135 to 230°C, fuel burnups from 8 to 34 GWD/MTU, and cover gas moisture levels from 0 to 40°C dew point. The tests were conducted in a 10^5 R/h gamma field.

From the scoping tests conducted at Pacific Northwest Laboratory (PNL) using spent fuel, it was determined that the initial rate of weight gain for spent fuel was more rapid than the rate for nonirradiated pellets even after correcting for the differences in geometric surface-area/volume ratios. Weight gain rates for spent fuel specimens were about 50 times greater than the rates before U_3O_8 powder formed for nonirradiated pellets.

The grinding of nonirradiated pellets into a powder caused less of an increase in the oxidation rate than predicted on the basis of linear increase with an increase in surface area. However, the rate of weight gain in nonirradiated pellets increased with decreasing grain size (increasing grain boundary surface area), indicating that the initial oxidation reaction occurs at intergranular sites in UO_2 . The rate of weight gain of nonirradiated pellets was also affected by temperature, storage time in air at ambient temperature prior to testing (aging time), density, oxidants formed by radiolysis, and to a lesser extent moisture level.

The period required to form U_3O_8 powder did not correlate with the rate of weight gain. For spent fuel, the formation of, apparently, U_3O_8 powder appeared at levels of approximately 2 to 3% increase in weight, whereas U_3O_8 powder formed at levels of weight gain as low as 0.05% for nonirradiated UO_2 pellets.

The imposition of an external gamma field did not affect the oxidation behavior of spent fuel when exposed to a continuous supply of fresh air from the hot cell atmosphere. Tests in a static atmosphere with an external gamma field are continuing to determine the effect, if any, of a gamma field. For nonirradiated pellets, U_3O_8 powder formation occurred after shorter periods at a given test temperature when the gamma field was present than without it. The increased rate of oxidation in a gamma field required a static atmosphere, and therefore was attributed to the formation of oxidizing products from air radiolysis.

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INTRODUCTION

Dry storage of spent light-water reactor (LWR) fuel at reactor sites is a licensed alternative in the United States to storage in pools. To date, however, licensed dry storage atmospheres have been restricted to inert gases. At sufficiently high temperatures, low-density respirable-size powders (U_3O_8 , UO_3) form during oxidation of spent fuel in air, and the volume expansion accompanying the oxidation can extend existing cladding breaches and, potentially, could lead to powder release through the breach. An understanding of spent fuel oxidation in air is required to 1) assess the fuel condition following an abnormal event during which a protective inert storage gas was replaced with air; 2) permit discontinuance of inert-gas purity monitoring during storage after the decay heat and fuel temperature has been sufficiently reduced; and 3) develop the basis for storage of spent fuel in air under certain temperature conditions. The first two reasons are associated with inert gas storage, but the latter relates to direct air storage. Technical bases for both scenarios are required. In addition, an understanding of fuel oxidation behavior is important to prevent fuel degradation from occurring during receiving, handling, and packaging operations.

To explore the oxidation behavior of UO_2 at anticipated air storage temperatures from 150 to 250°C, tests began in 1983 at Pacific Northwest Laboratory (PNL)^(a) as part of the Commercial Spent Fuel Management (CSFM) Program. When completed, the results from PNL tests, along with test results from other programs, will become the primary bases for developing recommended dry-storage temperature limits in air.

In this report, the final results of two series of spent fuel oxidation tests and the interim results of a third spent fuel test series are reported and discussed. In addition, the final results from a series of nonirradiated fuel tests are reported and discussed.

(a) Operated by Battelle Memorial Institute for the U.S. Department of Energy under Contract DE-AC06-76RLO 1830.

The purpose of the first spent fuel test was to determine if the oxidation behavior of nonirradiated fuel could be used to predict spent fuel oxidation. In this scoping test, pressurized-water reactor (PWR) fuel from one source reactor and nonirradiated fuel were oxidized in air at 200 and 230°C and gamma fields of 600 and 130,000 R/h.

The second spent fuel test was performed to determine if significant differences in oxidation behavior existed between spent fuel from different source reactors. A test series was conducted at PNL using fuel from three different PWR reactors in sealed ovens between 135 and 200°C in a low gamma field (500 to 1000 R/h). Samples included fuel with two levels of burnup from two of the reactors as well as nonirradiated fuel.

Results from the third spent fuel test, when completed, will be used to develop the design bases for storage of spent fuel in air and to determine the effect of abnormal storage conditions on the fuel. Results from previous PNL tests on spent fuel and nonirradiated fuel were used to identify the key variables for this long-term test (at least 3 years). A test matrix was developed using statistical experimental design principles. Tests are being conducted in sealed ovens and high gamma fields (10^5 R/h) to simulate conditions in a dry storage cask. Both PWR and BWR (boiling-water reactor) spent fuel with burnups from 8 to 34 GWd/MTU are being tested between 135 and 230°C and air moisture levels from 0 to 40°C dew points. Interim results up to about 4500 h of exposure are reported and discussed in this report. When these tests are completed, the final results will include data with exposure periods up to 22,000 h.

Nonirradiated fuel tests explored the oxidation behavior of UO_2 at temperatures between 150 and 250°C. These tests identified several important variables that can affect UO_2 oxidation. The final results of those tests are reported here.

DESCRIPTION OF PNL UO_2 FUEL OXIDATION TESTS

The general procedure followed for these tests was to place specimens contained in a sample boat inside an oven. The specimens were then heated to the test temperature and maintained at that temperature for some period of time. Periodically, the specimens were cooled, removed from the oven, weighed, and photographed. From these periodic weighings, the weight change was determined as a function of exposure period. Any weight gain was attributed to oxidation of the specimens. Occasionally, a small portion of the total specimen was removed for other analysis, for example, x-ray diffraction (XRD) or electron microscopy. In the sections below, specific details are reported about each test series.

SCOPING TESTS ON POINT BEACH SPENT FUEL IN AIR IN VENTILATED OVENS

Scoping oxidation tests were conducted at 200 and 230°C using spent fuel fragments and open-ended, 50-mm-long, fuel rod segments^(a) from the Point Beach PWR (Appendix A, Tables A.1 and A.4). The nominal burnup of the fuel was 29.5 GWd/MTU. The rod segments were included in the tests to evaluate the effect of decladding on fuel oxidation. Nonirradiated fuel pellets manufactured at PNL (White et al. 1983) were included in the tests as a control.

The spent fuel specimens were maintained within $\pm 3^\circ\text{C}$ of the nominal test temperature in air in ventilated ovens (White et al. 1983). One oven was located next to cesium-137 sources to generate a gamma field of about 150,000 R/h. The gamma field in the other oven was approximately 600 R/h.

Periodically, spent fuel specimens were removed from the ovens, weighed, and visually inspected through 4-ft-thick, lead-glass windows. Photographic and detailed examinations required transferring specimens to a separate hot cell where a periscope was located and, therefore, were performed less frequently than weighing. Small samples were removed from some of the oxidation test specimens for XRD analyses.

(a) Called "rod segments" throughout this report.

LOW-GAMMA SPENT FUEL TESTS IN HERMETICALLY SEALED OVENS

The next series of oxidation tests in air were conducted using spent fuel from three PWRs in hermetically sealed stainless steel ovens (Figure 1). Twelve ovens were located in two racks inside the hot cell. Each oven contained three shelves that held up to four fuel samples each. Thus, an oven could hold a maximum of 12 fuel samples in separate aluminum sample boats. Internal oven volume was 1.5 ft^3 (0.042 m^3). Temperature was monitored using chromel-alumel thermocouples positioned on each shelf between the sample boats. Internal oven volume was 1.5 ft^3 (0.042 m^3). Temperature was monitored using chromel-alumel thermocouples positioned on each shelf between the sample boats.

Nominal test temperatures were 135, 150, 165, 180, and 200°C (see Appendix B, Table B.1 for test matrix details) with a static cover gas having approximately a 30°C dew point. Fuel from three different PWRs was used (Table 1). Nonirradiated fuel pellets were included in the test matrix for comparison with previous tests. The hot cell background radiation and specimen self radiation (a total between 500 and 1000 R/h) were the only sources of gamma radiation.

FIGURE 1. Controlled-Atmosphere Oxidation Test Oven Rack

TABLE 1. Low-Gamma-Field Oxidation Test Fuels

<u>Source Reactor</u>	<u>Fuel Type</u>	<u>Burnup GWd/MTU^(a)</u>
Shippingport	PWR	23.7
Point Beach	PWR	29.5
H. B. Robinson	PWR	28.4 ^(b)

(a) Rod-average burnup unless otherwise noted.

(b) Gamma scan used to determine burnups of samples assumed a linear relation between cesium-137 counts and burnup (Barner 1985).

A series of three gas headers (cover gas inlet, cover gas outlet/vacuum, and cover gas sample) connected to the ovens permitted each oven to be operated with a controlled cover gas composition. Solenoid valves controlled gas flow to each oven. Cover gas was supplied via compressed gas cylinders located outside the hot cell. Pressurized breathing air was used for the low-gamma test series.

To obtain fuel for testing, 1.5- to 1.75-in.-long (3.8- to 4.4-cm) sections of a fuel rod were cut from a designated location using either a diamond wafering or aluminum oxide saw blade. Archive specimens and specimens for additional characterization (for example, ceramography and electron microscopy) were also removed and stored in air at approximately 30°C. A typical cutting diagram is depicted in Figure 2. Because the startup of an oven required specimens from several different fuel rods, sections were stored in labeled vials. When all materials for a particular oven were available, fuel was removed from the cladding by striking the cladding with a hammer and allowing the fragments to fall into a stainless steel collection pan. Fuel fines (less than No. 16 Tyler mesh screen) were sieved out from the larger fuel particles to avoid being confused with fine powder produced during fuel oxidation. The plus 16 mesh particles were placed in aluminum sample boats that had been previously weighed to ± 0.001 g. For each test temperature, PNL-produced, nonirradiated UO_2 pellets (White et al. 1983; White and Gilbert 1986) were included in each oven. All sample boats with fuel were weighed to within ± 0.001 g.

FIGURE 2. Typical Rod Cutting Diagram

Sample boats containing the fuel specimens were carefully placed onto oven shelf positions. The shelf position of each sample was invariant throughout the course of the tests. The oven door was sealed and the oven was evacuated to ~1-in. Hg (3.4 kPa) absolute pressure using a mechanical pump. After evacuation, compressed breathing air was routed from the inlet gas header and passed through a humidification tank where it became saturated at a dew point equal to hot cell ambient temperature (~30°C) as determined by a downstream moisture probe. Gas then flowed to the evacuated oven until ~32 in. Hg (109 kPa) absolute pressure was obtained. Evacuation and air backfilling were repeated to purge the oven chamber of possible gaseous contaminants and excess water vapor. The oven was then sealed by closing the inlet header valve, creating a static cover gas.

While the oven was heated to the desired testing temperature, periodic venting of the oven cover gas through the outlet/vacuum header cap screw was performed to prevent overpressurization and gasket blowout. Final oven pressure at test temperature was approximately 32 to 33 in. Hg (109 to 112 kPa) absolute pressure. The temperature of each oven shelf and the oven pressure were recorded daily during the tests.

Prior to oven shutdown, a sample of the cover gas was collected in an evacuated 500-mL bottle through the cover gas sample header. The samples were obtained as follows: the cover gas sample header was evacuated and backfilled

with purified helium seven times to remove possible contaminants; the header was under vacuum when the sample bottle valve and oven solenoid valve (which connected the header to the oven) were opened. The collected cover gas was analyzed with a mass spectrometer for CO₂, Ar, O₂, N₂, CO, He, H₂, and CH₄ (Appendix B, Table B.2). The cover gas was not sampled prior to oven startup.

After the samples cooled, sample boats were carefully removed from the oven shelves and weighed twice to within ± 0.001 g (± 0.01 g if total weight was 60 g or higher). Sample weight gain was attributed to oxidation of the fuel. Weight gain due to oxidation of the aluminum sample boats did not contribute significantly to the measured weight increase (see Appendix C). Samples were visually examined and periodically photographed through a hot cell periscope to detect powder formation. After an interim examination, the oven gasket was replaced, the samples were returned to their shelf positions, the oven door was sealed, and the oven was restarted as previously described.

HIGH-GAMMA SPENT FUEL TESTS IN HERMETICALLY SEALED OVENS

A series of spent fuel oxidation tests are being conducted in a high gamma field in the stainless steel ovens described in the last section. Nominal oven test temperatures are 135, 150, 170, 190, 210, and 230°C. A summary of the test matrix is given in Figure 3 and Appendix D, Table D.1. All 12 ovens (and all sample locations within each oven) are being used; thus, 144 samples comprise the high-gamma test matrix.

The BWR and PWR fuels from different reactors being tested in a high gamma field are described in Table 2. Gamma scans have been conducted on H. B. Robinson, Quad Cities-1, and Monticello fuel rod segments using a germanium-lithium gamma-ray detector. The burnup levels of samples cut from these rods at various locations were estimated by assuming a linear relation between cesium-137 counts (from the gamma scan) and burnup (Barner 1985). The burnup levels of samples cut from the Point Beach and Shippingport rods were assumed to be the average burnup (based on power history) for the entire rod as shown in Table 2.

To simulate the gamma field under storage conditions, a CsCl gamma source capsule was placed in front of each oven door while the samples were at test

Temperature, °C		135	150	170	190	210	230
Dew Point, °C	0	1	1		1		
	20		1	2	1	1	1
	40	1	1		1		

FIGURE 3. Summary of High-Gamma-Field (10^5 R/h) Oxidation Test Matrix (numbers refer to the number of ovens at the stated test conditions)

temperature. Dose rates of the sources are approximately 1.3×10^5 ($\pm 14\%$) R/h at 12 in., which is approximately the distance of the samples to the source. The purpose of the imposed gamma field was to simulate the gamma radiation in a storage cask.

In order to perform the tests at three different moisture levels (0°C, 20°C, and 40°C dew points) under a static cover gas, modifications to the cover gas piping used for the low-gamma test series were made, and changes in the final backfilling procedure were necessary. Commercial-grade compressed dry air was used as the gas source.

The modified system allows cover gas to be routed simultaneously through a humidification tank (where moisture evaporates into the gas stream) and a humidification tank bypass. The two gas streams (one dry, the other at or near saturation) mix and pass across a moisture probe before entering an oven. Thus, by adjustment of the flow rates of the two streams with separate rotameters and heat input to the humidification tank (above ambient dew points only), controlled moisture levels are obtained.

TABLE 2. High-Gamma-Field Oxidation Test Fuels

Source Reactor	Rod Identification	Fuel Type	Burnup Gwd/MTU ^(a)
Shippingport		PWR	23.7 ^(b)
Point Beach		PWR	29.5 ^(b)
H. B. Robinson		PWR	28.4 ^(c)
Quad Cities - 1			
	1A01-1	BWR	22.6 ^(c)
	1A01-2	BWR	7.5 ^(c)
	1A03-3	BWR	12.0 ^(c)
	1B02-4	BWR	7.5 ^(c)
	1C02-1	BWR	15.9 ^(c)
	1C02-4	BWR	22.6 ^(c)
	DTA0103	BWR	16.6 ^(c)
Monticello			
	0A03-4	BWR	28.7 ^(c)
	0A08-4	BWR	28.7 ^(c)
	5D18-3	BWR	28.7 ^(c)

- (a) Average burnup of rod or rod segment unless otherwise noted.
- (b) Estimated average burnup based on power history.
- (c) Gamma scan used to determine burnups of samples assumed a linear relation between cesium-137 counts and burnup (Barner 1985).

Samples were selected from designated fuel rod locations prior to the cutting in order to provide a range of burnups and fuel type (from both BWR and PWRs) for each oven. To provide a statistical estimate of within-oven variability, replicate samples (same burnup and fuel type) were placed in an oven when possible. Two ovens were operated at identical conditions (170°C, 20°C dew point) to estimate between-oven variability.

Because the startup of an oven required specimens from several different fuel rods, freshly cut segments were stored within 5 days after cutting in a nitrogen atmosphere until all samples for a particular oven were prepared. Eleven spent fuel specimens were removed from nitrogen storage for each oven, declad, and prepared for testing as previously described in the low-gamma test section. Typical size of the fragments prior to testing was ~5 to 10 mm on edge (1/4 to 1/3 of a pellet) for fuel with relatively low burnup

(<20 GWd/MTU), 4 to 6 mm on edge for fuel with intermediate burnup (~20 to 25 GWd/MTU), and 4 mm or less for high-burnup fuel (>25 GWd/MTU). The lower-burnup fuel was easier to remove from the cladding, and thus the larger fragment sizes may be partly due to less fracturing as a result of decladding but may also be due to the lower number of thermal cycles or power level (which causes fuel fracturing) encountered during reactor operation. Well-characterized, PNL-produced, nonirradiated UO_2 pellets were added (typically three) to the twelfth sample boat in each oven. All sample boats with fuel were weighed twice to within ± 0.001 g (or ± 0.01 g if total weight was 60 g or over). Differences in weight between weighings of a single sample were typically 0.001 to 0.002 g. Initial spent fuel weights ranged from 18 to 46 g depending on fuel type and length of the rod segment. The initial weight of the nonirradiated pellets was ~16 g (~5.3 g pellet).

With the exception of the final backfill, oven test startup was similar to that previously described for the low-gamma tests. On the final backfill, the cover gas was vented through a loosened cap screw located on the outlet/vacuum header. Once the correct dew point was obtained (usually to within $\pm 2^\circ\text{C}$ for 0°C and 20°C dew points), the cover gas was allowed to flow through the test oven for a minimum of three oven-volume exchanges. The oven was then sealed by closing the inlet header valve and cap screw.

For a dew point of 40°C ($\pm 5^\circ\text{C}$), cover gas flow was adjusted in a manner similar to the 0 or 20°C dew points, but the cover gas was vented through an empty oven (with the door open). Simultaneously closing the inlet header valve to the empty oven and opening the inlet header valve to the evacuated oven backfilled the oven with a cover gas of the desired dew point. This prevented possible condensation inside the sealed test oven that could result from using the continuous flow method in the test oven. Dew points above cell ambient temperature required heating the humidification tank and lines to the ovens.

The period of time that an oven was at test temperature before shutdown for an interim weighing was dependent on the temperature, with the higher temperature tests being shut down more frequently, particularly during the early stages of the tests.

A cover gas sample was taken prior to oven shutdown and analyzed for CO_2 , Ar, O_2 , N_2 , CO, He, H_2 , CH_4 , and NO_x (Appendix D, Table D.3). During the most recent interim examinations, the samples were also analyzed for radiolysis products (Xe, Kr). A pretest cover gas analysis was taken each time a dry air cylinder was replaced so that the starting composition of the gas was known (Appendix D, Table D.2).

After each oven cooled, the specimens were removed and weighed twice to within ± 0.001 g. Specimens were visually examined and periodically photographed and then returned to the oven. The ovens were restarted as previously described.

NONIRRADIATED UO_2 PELLET TESTS

Tests using PNL-produced, nonirradiated UO_2 pellets involved placing bare pellets in an oven with an air cover gas. The pellets tested at temperatures from 150 to 250°C were periodically weighed to determine the kinetics of oxidation and examined to detect physical changes, such as the formation of powders. Selected specimens were subjected to x-ray examination to determine the composition and structure of these powders. Above 250°C, continuous weight measurements using a recording balance were taken to monitor the rapid rate of weight gain. Other variables besides temperature were also examined. These variables are discussed below.

The quality of the air was varied: atmospheric air (Appendix E), air with 1 vol% NO_2 gas to synthesize an oxidizing radiolysis product (Appendix F), and air with controlled moisture content (Appendix G). The oxidation behavior of nonirradiated UO_2 pellets containing up to 10 wt% gadolinia was also investigated (Appendix E).

Some nonirradiated pellets were also subjected to a 200,000 R/h cobalt-60 gamma source during oxidation in air to evaluate the effects of radiolysis (to simulate storage conditions) of the cover gas on the oxidation behavior (Appendix H).

The period that the hydrogen-sintered pellets had been exposed to ambient atmosphere prior to testing (aging time) was investigated to resolve

differences in oxidation behavior between pellets subjected to testing immediately after a hydrogen reduction and pellets stored in ambient atmospheric air before testing. These data are presented in Appendix E.

Tests were performed on nonirradiated pellets and mechanically ground pellets to determine the effect of surface area on oxidation behavior. The results of these tests are presented in Appendix I.

In order to study the relationship between oxidation behavior and pellet character and to attempt to resolve differences in published data, nonirradiated pellets from the United Kingdom Central Electricity Generating Board - Berkeley Nuclear Laboratory (UKCEGB-BNL) and the Chalk River Nuclear Laboratories (CRNL) were tested in common ovens with PWR-type pellets manufactured at PNL. Specimen characterization and test results are presented in Appendix J. Air oxidation tests were performed at 200 and 220°C. Four of the UO_2 pellets obtained from each of the CRNL, UKCEGB-BNL, and PNL oxidation test programs were reduced in hydrogen at 1000°C for 2 h to bring the pellets to an initial common state at the beginning of the oxidation tests. Four pellets (two reduced, two as-received) from each laboratory were tested at 200°C and four at 220°C in air at 0°C dew point. The specimens were periodically removed from the ovens for weighing and visual inspection. Other experimental procedures were reported in a previous publication (White et al. 1983).

In the above comparison test, the immersion density of each pellet was measured before test startup, and a pellet from each laboratory was destructively examined for grain size and microstructural evaluation. The grain size of the three groups of pellets varied from an average of 4.9 μm for PNL pellets and 9.8 μm for CRNL pellets to 22.6 μm for UKCEGB-BNL pellets as determined by the grain intercept method described in Section 7 of ASTM standard E-112 (1973). Ceramographs of the three pellet types are shown in Figures 4 through 6. To compensate for the effect of the differences in pellet size, the data were reduced into a weight gain parameter normalized to the geometric external surface area of the pellet (mg/cm^2). This parameter characterized the starting geometric surface area only and did not account for any internal surfaces or changes in pellet surface area during oxidation testing.

FIGURE 4. Micrograph of Nonirradiated UO_2 PNL Pellet

FIGURE 5. Micrograph of Nonirradiated UO_2 CRNL Pellet

FIGURE 6. Micrograph of Nonirradiated UO₂ UKCEGB-BNL Pellet

Temperature-change tests were performed on nonirradiated UO₂ pellets to generate data for comparing oxidation behavior under changing temperature conditions with the predictions of cumulative damage models. These results are presented in Appendix K.

RESULTS AND DISCUSSION

The results of three oxidation tests using spent fuel are discussed in chronological order in the following sections. Results from the first test influenced the variables studied in the second, and the results of the second test influenced the third test matrix. A separate series of tests using non-irradiated pellets were also performed, and are discussed after the spent fuel tests.

SCOPING TESTS ON POINT BEACH SPENT FUEL IN VENTILATED OVENS

Point Beach spent fuel fragments and rod segments, along with nonirradiated pellets, were tested in air in ventilated ovens in gamma fields of 600 R/h at 200 and 230°C. A separate set of specimens at 230°C was also tested in a field of 150,000 R/h. The initial matrix included a test at 200°C and 150,000 R/h, but it was terminated after 185 h because of an oven failure. The appearance of fragments prior to oxidation is shown in Figure 7. The fragments

FIGURE 7. Point Beach Spent Fuel Prior to Oxidation Test (scale in inches)

have edge dimensions of 3 to 5 mm. After 5400 h, these test were terminated. The results are shown in Figures 8 and 9 for the 200 and 230°C tests, respectively. The data are tabulated in Appendix A.

Results at 200°C

At 200°C, the spent fuel fragments and rod segments exhibited an initial period of rapid oxidation, followed by a transition period where the rate of oxidation decreased. In contrast, measurable oxidation of the nonirradiated pellets did not begin until 1700 h, at which point the oxidation was rapid up to 2700 h. Between 2700 and 5400 h, very little oxidation of the nonirradiated pellets occurred. At the conclusion of these tests, the spent fuel fragments and rod segments had gained 1.9 and 1.8 wt%, respectively, and the nonirradiated pellets had gained 0.8 wt%.

The spent fuel fragments and rod segments oxidized at nearly the same rate up to about 2000 h. Above 2000 h, the oxidation of the fuel in the rod

FIGURE 8. Comparison of Oxidation of Point Beach PWR Spent Fuel Fragments and Rod Segments with Nonirradiated Pellets at 200°C (Tables A.2 and A.3)

FIGURE 9. Comparison of Oxidation of Point Beach PWR Spent Fuel Fragments and Rod Segments with Nonirradiated Pellets at 230°C (Tables A.5 and A.6)

segments was slightly lower than the fragments. Considering a relative standard deviation (RSD) of 25 to 27%,^(a) this difference is not considered significant.

Particulates less than 1 mm in size were first observed among the spent fuel fragment and rod segment specimens at the same time (2942 h); however, particulates were not observed in the nonirradiated pellet specimens at any time. Oxidation of nonirradiated pellets produced a fine powder. The spent fuel particulates (Figure 10) are easily distinguished from the 3- to 5-mm fragments.

(a) This RSD was determined during spent and nonirradiated fuel tests. The RSD determination is discussed in the High-Gamma Spent Fuel and Nonirradiated Results and Discussion sections.

FIGURE 10. Fragments and Particulates of Point Beach Spent Fuel After 2942 h at 200°C in a Gamma Field of 600 R/h (scale in inches)

X-ray diffraction measurements on fragments removed before particulate formation (at 1422 h) revealed broad and weak lines characteristic of an oxygen-rich tetragonal UO_2 phase. X-ray diffraction measurements were not made on the particulates.

Results at 230°C

At 230°C, the spent fuel fragments and rod segments oxidized rapidly up to 1000 h after which the rate of oxidation continuously decreased and reached a plateau after 1700 h (Figure 9). The fragments tested in the low gamma field oxidized at a faster rate and to a higher level (3.3 wt% after 5400 h) than the fragments tested in the high gamma field (2.5 wt% after 5400 h). Thomas, McCarthy, and Gilbert (1986) identified the fragments as U_4O_{9+x} .

The fuel in rod segments oxidized at a lower rate and had less total oxidation than the fragments. The fuel in the rod segment in the high gamma field had gained 2.0 wt% after 5419 h. The rod segment tested in the low gamma field was terminated after 836 h and a weight gain of 1.8%.

The nonirradiated pellets did not show significant weight gain until after 1200 h in the low gamma field and after 550 h in the high gamma field. Once oxidation began, the rate continually increased until the test was terminated. The nonirradiated pellets tested in the high gamma field oxidized more rapidly than those in the low gamma field.

Particulates less than 1 mm were detected among the fragments after 2835 h in the low gamma field (Figure 11). These particulates were identified by XRD as primarily U_4O_9 with a trace of U_3O_7 . No evidence of U_3O_8 was found in these particulates. At 996 h, fragments were characterized by broad and weak XRD peaks as oxygen-rich tetragonal UO_2 . No particles were found among the fragments tested in the high gamma field or among the rod segments in either gamma field.

U_3O_8 powder (identified by XRD) was found among the nonirradiated pellets at 1000 and 550 h in the low- and high-gamma-field tests, respectively.

FIGURE 11. Fragments and Particulates of Point Beach Spent Fuel After 2835 h at 230°C in a Gamma Field of 600 R/h (scale in inches)

Comparison of Spent Fuel and Nonirradiated Pellets

The oxidation behavior of spent fuel was dramatically different from that of nonirradiated pellets (Figures 8 and 9). At both temperatures, the initial rate of oxidation of spent fuel was significantly higher than for the nonirradiated pellets. For example, at 230°C, the rate differed by a factor of 50. This difference cannot be totally attributed to differences in surface area or grain size. Assuming spent fuel fragments were 3-mm cubes and neglecting surface roughness, the geometric surface-area/volume ratio of the spent fuel fragments was only two to four times greater than for nonirradiated pellets. Assuming a linear relationship between surface-area/volume ratio and oxidation rate, the surface-area/volume ratio difference between spent fuel fragments and nonirradiated pellets accounts for only a difference in rate of oxidation of two to four times, not the 50 times observed experimentally. Surface roughness would have to be determined before the effect of surface area can be completely ruled out; this has not been done to date.

The effect of fission products on UO_2 may partially account for the difference between spent and nonirradiated fuel. During irradiation, fission products are produced in the UO_2 fuel. The fission products affect the thermodynamic and chemical properties of spent fuel. Fission products occupying lattice and interstitial sites can alter the potential for oxidation of UO_2 by generating defects and changing the uranium valence to maintain charge neutrality. Smith (1960) has demonstrated that the oxidation behavior of UO_2 doped with simulated fission products is different from that of undoped UO_2 . Thomas, McCarthy, and Gilbert (1986) has shown the microstructure of irradiated UO_2 changes to accommodate fission products by forming bubbles at the grain boundary.

From this scoping test, it was determined that the oxidation behavior of spent fuel cannot be predicted from nonirradiated pellet oxidation. A small portion of the difference between spent fuel and nonirradiated pellet behavior could be attributed to different geometric surface areas. The major behavioral differences probably lie in the fact that during irradiation, the thermodynamic, chemical, and physical properties, and microstructure of UO_2 change

to the point where the spent fuel no longer behaves like nonirradiated UO_2 . These changes in UO_2 properties need to be examined and related to spent fuel oxidation behavior.

Effects of Gamma Radiation and Cladding

Higher gamma radiation apparently reduced the rate and total amount of oxidation and increased the time for particle formation of spent fuel fragments and rod segments at 230°C . However, considering an RSD of 25 to 27%, this difference cannot be considered significant. In contrast, the high gamma radiation appears to enhance the oxidation of nonirradiated pellets.

At 200°C , the fuel in the rod segments and fragments exhibited similar oxidation behavior. However, the oxidation of fragments at 230°C was higher than that of the fuel in the rod segments. This difference is not considered significant when the RSD is taken into account.

The rate of weight gain (%/h) of the rod segments was less than the rate of the bare spent fuel fragments by a factor of one-third to one-half at 230°C (Figure 9). Partial protection of the fragmented fuel surfaces by the cladding might limit oxygen ingress and reduce the observed rate of weight gain. However, because of ineffective protection by the cladding in the 200°C test, this explanation does not appear to be adequate. Other unknown factors may be important.

Mechanism for Spent Fuel Oxidation

Wheeler (1983) predicted that the oxidation of UO_2 would be diffusion controlled. Bulk diffusion-controlled oxidation would result in parabolic behavior with a slope = 0.5 when the log weight gain is shown versus log time. Experimental results from the oxidation of Point Beach spent fuel at a temperature of 230°C (Figure 12) indicate an initial linear rate of weight gain with time and a slope of 1, followed by a transition period of decreasing slope. Beyond 300 h, the rate of weight gain is again constant with time with slope much less than 0.5. Consequently, the results do not support bulk diffusion-controlled oxidation.

The initial linear region with slope of 1 suggests surface-controlled reactions. The linear period beyond 300 h is hypothesized to be the result of

FIGURE 12. Time Dependence for Point Beach Spent Fuel Oxidation in Air at 230°C (Table A.5, Specimen 1D, 29.5 GWd/MTU burnup)

a U_3O_7 or U_3O_8 protective layer forming on exposed surfaces. U_3O_7 and U_3O_8 films on UO_2 are expected to be protective, but the effectiveness of these films as a diffusion barrier is not well characterized (Colmenares 1975, 1985). The oxidation rate becomes slow prior to complete oxidation to an average composition of U_3O_8 , indicating that the oxide coating being formed (perhaps U_3O_7 or U_3O_8) is very protective or that an induction time is required for the formation of U_3O_8 . Auger electron spectroscopy, transmission electron microscopy (TEM), and XRD analysis are planned to help determine the oxidation mechanisms of spent fuel.

LOW-GAMMA SPENT FUEL TESTS IN HERMETICALLY SEALED OVENS

Results of tests with spent fuel fragments in air at temperatures between 135 and 200°C are presented in Figure 13 and Appendix B, Tables B.3 through B.7. The test variables included five temperatures, four fuel types (Table 3), two different burnup levels for the Point Beach and H. B. Robinson fuels, and two cumulative exposure times-at-temperature. All spent fuels were PWR-type, obtained from three different reactors.

FIGURE 13. Oxidation of PWR Spent Fuel Fragments and UO_2 Pellets at 135 to 200°C in a Gamma Field of 500 to 1000 R/h

TABLE 3. Sources of Spent Fuel Fragments for Low-Gamma Tests

Designation	Reactor	Burnup, Gwd/MTU
PB 20	Point Beach	20
PB 30	Point Beach	29.5
HBR 22	H. B. Robinson	21-23
HBR 31	H. B. Robinson	31
SP	Shippingport	23.7 (avg) 34 (peak)
NI	Not irradiated	0

Powder, or particulates, was not observed to have formed on any of the spent fuel samples after approximately 5000 h of testing. The weight gain levels for all spent fuels were about 2.3 wt% at a temperature of 200°C. Powder was observed to have formed on the PNL-produced, nonirradiated fuel pellets prior to 5070 h of exposure in air at a temperature of 200°C, with a weight gain of ~0.5 wt%.

The results (Figure 13) indicate a general increase in weight gain with both increasing temperature (except at 180°C) and increasing exposure period. The lower than expected weight gains at a temperature of 180°C may be due to differences in the sample preparation procedure, whereas the procedure for specimen preparation involved removing the pellet fragments from the cladding and promptly initiating the 135, 150, 165, and 200°C tests. Specimen loading into the 180°C oven was delayed for approximately 6 weeks because of hot cell operational circumstances. A thin protective coating consisting of UO_3 hydrate is postulated to have formed on the surface of the declad specimens while they were stored in air at cell ambient temperature; effects on oxidation behavior of fuel exposure to ambient temperature conditions consistent with this postulate have previously been reported (Wadsten 1977; Gilbert, White, and Knox 1984; Hastings et al. 1984).

A statistical analysis on weight percent gain per hour data was performed to determine the significance level of three factors: temperature, fuel-type/burnup combination, and exposure period. The fuel-type/burnup combination refers to a specimen from one source reactor at one burnup level. The significance of two-way interactions between these three factors was also determined. For this analysis, the importance of a factor increases as the significance level decreases; a significance level below 0.100 was considered an important effect. The results (Table 4) indicate a significant linear dependence on temperature and a significance of fuel-type/burnup combinations and exposure period. Fuel-type/burnup combination had the most significant effect compared with the other two factors. A quadratic dependence of temperature was not significant. The quadratic dependence of temperature could be masked by the operational difficulties with the 180°C tests that were discussed above. There

TABLE 4. Results of Split-Split Plot Analysis of Variance
for Low-Gamma (500 to 1000 R/h) Air Oxidation
Data Between 135 and 200°C

Effect	Significance Level ^(a)
Temperature	
Linear effect	0.066
Quadratic effect	0.782 ^(b)
Fuel-Type/Burnup Combinations ^(c)	<0.0001
Exposure Periods	0.009
Two-way Interactions:	
Fuel Type * Linear Temperature	0.005
Fuel Type * Quadratic Temperature	0.061
Exposure Period * Linear Temperature	0.016
Exposure Period * Quadratic Temperature	0.809
Exposure Period * Fuel Type	0.006

- (a) The smaller the significance level (p-value), the more significant the effect. For the purpose of this analysis, a significance level below 0.10 was considered to be a significant effect.
- (b) Operational problems with 180°C test samples (described in text) may mask significance of quadratic nature of temperature effect.
- (c) The fuel-type/burnup combinations included in the analysis were NI, HBR 31, PB 30, PB 20, and SP (see Table 3 for definitions). The HBR 22 type was omitted due to lack of data at two temperatures.

were also several significant two-way interactions between the three factors, which indicates that the effects of each of the factors depend on the levels of the others.

A preliminary evaluation to determine possible causes for the differences in weight gain among the three fuel types indicated that a positive correlation between the level of weight gain and extent of microporosity may exist. Differences in microporosity among specimens may be attributable to differences in the formation of fission gas bubbles during reactor operation and/or difference in fuel manufacturing methods. During the initial stages of oxidation, Shippingport spent fuel showed consistently large weight gains when compared with

specimens from other fuels with comparable burnup (Figure 13). The microstructure of Shippingport spent fuel showed significant microporosity (Figure 14). This microporosity may provide interconnected paths for air to oxidize interior surfaces of spent fuel fragments. Ceramographic studies of Shippingport fuel samples taken from positions adjacent to the oxidation specimens are needed to quantify the microporosity and volumetric ratio of interconnected pores.

Oxidation test specimens and ceramographic specimens were prepared from adjacent locations near the center and at the end of H. B. Robinson spent fuel rod N-9. The weight gain of specimen HBR 31 (Figure 13) prepared from near the fuel rod centerline high-power/high-burnup location (highest irradiation temperature is 1000°) was larger than for specimen HBR 22 prepared from moderate-power/moderate-burnup location (near the end of the rod) at 150°C oxidation test temperature. The microstructure of the ceramographic specimens showed more microporosity for the centerline region of specimen HBR 31 (Figure 15b) than for the end region (Figure 15a), which is an indication of a correlation

FIGURE 14. Microstructure of Shippingport Fuel (Rod 4) at 1/2 Radius (Bradley et al. 1981)

a) Near Moderate-Power/Moderate-Burnup Position (22 GWd/MTU)
(as-polished transverse sample N-9B-0; Barner 1985)

b) Near High-Power/High-Burnup Position (30 GWd/MTU)
(as-polished transverse sample N-9C-C; Barner 1985)

FIGURE 15. Fuel-Pellet Centerline Microstructure of H. B. Robinson Spent Fuel Near Moderate-Power and High-Power Positions

between microporosity and oxidation behavior. The possibility of other differences in fuel characteristics, e.g., fission product concentrations, resulting from differences in burnup cannot be ruled out. However, significant radial variability in microporosity was observed in transverse ceramographs of H. B. Robinson fuel rod N-9 (Barner 1985). As a result, in most cases negligible differences in the average rate of weight gain between H. B. Robinson specimens would be expected because of the relatively large volume of samples used in the tests. This expectation agrees with the test results of H. B. Robinson fuel samples exposed at temperatures of 180 and 200°C; the H. B. Robinson fuel tested at 150°C may be an experimental anomaly. Ceramographic and microscopic studies of specimens taken from positions adjacent to the oxidation specimens may clarify the effect of microstructure on oxidation of spent fuel.

HIGH-GAMMA SPENT FUEL TESTS IN HERMETICALLY SEALED OVENS

Interim results are reported for these tests following exposure periods of up to 4500 h. When the tests are completed, the final results will include exposure times of 22,000 h. It is difficult to evaluate statistically designed experiments until all the data are available. The interim results include less than 20% of the planned exposure time. Therefore, only general trends will be discussed below with emphasis on the highest temperature data where most of the weight gain has occurred.

Before discussing the results, the experimental error will be addressed. The experiment design provided for estimation of the experimental error variability. There are two main classes of experimental variability to be considered--"within-oven variability" and "between-oven variability." Within-oven variability is partially attributed to differences in temperature distributions between oven shelves and between specimen locations for a given shelf, and partially attributed to differences in specimens of the same fuel type and burnup, e.g., burnup uncertainties, microstructure, fabrication history, and fission product concentration. Between-oven variability is due to differences in ovens, differences in gamma field intensity for each oven, and other contributions related to potential oven differences. Preliminary analyses indicate that between-oven variability is small compared to within-oven variability.

Within-oven RSDs of weight percent gain data varied from less than 1% to 86% with the majority of RSDs below 27%. The RSDs are computed from sets of two or three replicates or near replicates (specimens of the same fuel type with the same or nearly the same burnup from the same oven).

Oxidation of PWR Spent Fuels

Weight gain behavior for PWR spent fuels at 230°C is shown in Figure 16. Below 1000 h, the weight gain behavior of the PWR fuels was different. The initial rate of weight gain varied up to a factor of 5 (Figure 16), but the overall weight gains for all three fuel types were similar after 1000 h. Shippingport fuel in almost all cases had the highest initial rate of weight gain among the spent fuel samples for a given time and temperature (Appendix D, Table D.4). For example, at 230°C and 752 h, Shippingport sample 1B-1 gained weight at a rate of 2.98×10^{-3} wt%/h, which was a factor of 3 greater than the Point Beach fuel and 6 to 14 times that of H. B. Robinson fuel. These results are consistent with results of the low-gamma tests for exposure times between

FIGURE 16. Weight Gain Behavior of PWR Fuels Tested at 230°C, 20°C Dew Point

1518 and 1705 h (Figure 13). Microstructures of Shippingport spent fuel show significant microporosity (Figure 14), which may account for the increased rate of oxidation.

Between 1000 and 2000 h, the rate of weight gain was not as rapid as the initial rates. The level of weight gain increased to approximately 3 wt% at the 2000 h mark. For the exposure time period between 2000 and 4000 h, the rate of weight gain appeared to follow a behavior indicative of a diffusion-controlled reaction.

In all PWR fuels tested, the level of weight gain after 4000 h of exposure exceeded 4 wt% (a 4% gain corresponds to complete conversion of UO_2 to U_3O_8). The total weight gain was attributed to oxidation of the specimen because no significant weight change resulted for an empty aluminum sample boat after approximately 2200 h of exposure at 230°C test conditions. Also, no visible degradation of the sample boats was observed. Apparently, the fuel proceeded to a higher oxidation state, possibly UO_3 .

FIGURE 17. Weight Gain Behavior of BWR Fuels Tested at 230°C, 20°C Dew Point

Oxidation of BWR Spent Fuels

The weight gain behavior at 230°C of BWR spent fuels is shown in Figure 17. The multiple points in Figure 17 represent different burnup levels. In general, BWR fuels showed an overall weight gain behavior that appeared to follow parabolic kinetics, suggesting a diffusion-controlled oxidation process (Aronson, Roof, and Belle 1957). However, below 1100 h, there was some indication of a linear dependence of weight gain with time. A linear dependence of weight gain with exposure time has been predicted for surface-controlled reactions under isothermal conditions and constant surface area (Aronson, Roof, and Belle 1957). Between 1100 and 4000 h, the fuels exhibited approximate parabolic behavior suggesting a bulk diffusion-controlled reaction to be rate controlling. Similar to the PWR fuels, the level of weight gain of the BWR fuels exceeded 4 wt% after 4000 h.

The weight gain behavior for nonirradiated pellets tested at 230°C was sigmoidal with time (Figure 18). The sigmoidal shape of weight gain with exposure time for nonirradiated UO_2 pellets has been observed at 250°C by

FIGURE 18. Weight Gain Behavior of PNL-Produced, Nonirradiated Pellets at 230°C, 20°C Dew Point

Simpson and Wood (1983). The weight gain of the nonirradiated pellets was nearly 6 wt%, corresponding to nearly complete conversion to UO_3 .

Effect of Burnup on Spent Fuel Oxidation

For the BWR fuels, weight gains of up to 5% were essentially independent of burnup from 8 to 34 GWd/MTU at a temperature of 230°C (Figure 19). This is consistent with results from analysis (Gilbert, White, and Knox 1984) of oxidation data corresponding to different burnup levels (Harrison, Padgett, and Scott 1967). Although the PWR fuels in the 230°C test appeared to follow the same trend, the range of burnups (24 to 30 GWd/MTU) is too narrow to draw any conclusions at this time.

Because of operational difficulties, pretest 230°C specimens were not photographed and visual observations of 230°C test specimens were not made until 1664 h. However, specimens tested at 135°C were similar with regard to source reactor and burnup as specimens tested at 230°C. Because powder was not

FIGURE 19. Weight Gain as a Function of Burnup Level for BWR Fuels Tested at 230°C. (Data points at 24.2 and 33.9 GWd/MTU burnup after 3961.5 h of exposure are omitted because fuel spillage was suspected.)

observed among the specimens tested at 135°C after 3192 h (Figure 20), these specimens are used to photographically approximate the pretest condition of specimens tested at 230°C.

In general, the visual examinations of BWR fuels revealed semiglossy sharp-edged fragments prior to powder formation. However, BWR fuels at approximately 34 GWd/MTU exhibited a metallic luster. Powder formation was usually accompanied by a transition of the fragments to a dull sheen and the disappearance of the sharp edges when large quantities of powder were present (20 to 30%). Such a fragment, removed for future TEM analysis, easily broke into several pieces during handling. Deep open cracks to multiple hairline cracks in the fragments were usually present when powder had formed.

Examination of BWR samples after exposure at 230°C revealed a trend of increasing volume of powder with decreasing burnup. The estimated amounts of powder formed as a function of time are given in Table 5. After 1664 h of exposure, an estimated 95% (by volume) of the nonirradiated pellets (0 GWd/MTU burnup) were converted to powder; 10 to 15% powder was observed on fragments at 8.0 GWd/MTU burnup; and less than 5% powder was observed at 12.0 GWd/MTU burnup. The time to powder formation for the specimens with burnups of 12.0 GWd/MTU or less was therefore below 1664 h. Powder was starting to form on the fuel fragments at 17 GWd/MTU burnup, and no powder was observed on

FIGURE 20. BWR Spent Fuel Exposed 3192 h at 135°C in Air (40°C dew point) Used to Approximate Pretest Condition of Specimens Oxidized at 230°C. Values below photos are burnup levels in GWd/MTU.

TABLE 5. Estimated Volume Percent Powder Formed on Bare Fuel Specimens at 230°C as a Function of Time from Visual Examinations

Sample ID	Fuel Type	Burnup, Gwd/MTU	Exposure Time, h					
			1664	1997	2502	3223	3962	4617
12B-12	NI	0.0	95	95	100	100	100	100
12B-6	BWR	8.0	10-15	10-15	10-15	15-20	15-20	20-30
12B-7	BWR	8.0	10-15	10-15	10-15	15-20	15-20	20-30
12B-4	BWR	12.0	5	5	5	5	5	20-30
12B-8	BWR	17.2	0	0	<1	<1	<1	5
12B-1	PWR	23.7	0	0	0	0	0	0
12B-9	BWR	24.2	0	0	0	0	0	<1
12B-3	PWR	26.8	0	0	0	0	0	0
12B-2	PWR	29.5	0	0	0	0	0	0
12B-11	BWR	33.9	0	0	0	0	0	<1
12B-10	BWR	34.4	0	0	0	0	0	<1

samples at higher burnups. Examination of specimens after 1997 h of exposure (Figure 21) showed no significant changes in the state of the fuel specimens observed at 1664 h.

The powder volume of BWR specimens which previously had powder present after 1664 h of exposure increased with increasing exposure time. After 4617 h of exposure at 230°C (Figure 22), powder was observed in all the BWR fuel

FIGURE 21. BWR Spent Fuel Exposed 1997 h at 230°C in Air (20°C dew point). Values below photos are burnup values in Gwd/MTU; arrows in photos show location of powder.

specimens. The same specimens are shown in Figures 21 and 22. These results show that for the BWR fuel tested, the time to powder formation on bare fuel fragments with burnup levels from 17 to 34 GWd/MTU increases with increasing burnup. This trend also appears to be valid below burnups of 17 GWd/MTU assuming that a greater volume of powder present in a specimen indicates a lower time to powder formation.

The increase of time to powder formation with burnup was proposed by Gilbert et al. (1983) for a variety of LWR fuels under different test conditions between 229 and 283°C. Einziger and Strain (1986) observed a similar trend with Turkey Point nonirradiated pellets and bare Turkey Point PWR spent fuel fragments at 295°C.

Powder has not been observed on any of the PWR fuel specimens after 4617 h of exposure, the period required for powder formation on the PWR fuels appears to be longer than for the BWR fuels. This is not due to burnup differences between the BWR and PWR fuels because the BWR burnup range of 8 to 34.4 GWd/MTU brackets the narrower PWR burnup range.

NONIRRADIATED UO₂ PELLET TESTS

Initial tests on PNL-produced, nonirradiated UO₂ pellets were started at 300 and 345°C with a continuous weighing system (White et al. 1983); however, the oxidation rates were so rapid (Figure 23) that lower temperature tests were

FIGURE 22. BWR Spent Fuel Exposed 4500 h at 230°C in Air (20°C dew point). Values below photos are burnup levels in GWd/MTU; arrows in photos show location of powder.

FIGURE 23. Continuous Weight Gain Data of Nonirradiated UO_2 in Air

required for evaluating extended storage periods. Furthermore, the continuous weighing system prevented inspection of the specimens for powder formation and limited the number of specimens that could be tested. A weight gain of 4%, reached in the 345°C test, corresponds to 100% conversion of UO_2 to U_3O_8 powder.

The next set of tests was performed between 150 and 250°C with interruption for weight measurements. The results of these tests are shown in Figure 24. The data were found to show pellet-to-pellet variability of up to 25% (RSD) for tests using multiple pellets (test temperatures of 150, 180, 200, and 230°C). The results of RSDs computed to measure this variation are summarized in Table 6. Statistical regression techniques were utilized to determine whether pellet density, surface area, or surface condition (as-sintered versus freshly ground) explained (or were causes of) any of the pellet-to-pellet weight gain variation. The regression techniques were applied separately to the data sets for each period for each temperature to allow for the possibility that a given variable might explain differences in weight gain among pellets for certain times (stages of oxidation) but not others.

FIGURE 24. Nonirradiated UO_2 Oxidation Behavior Between 180 and 250°C (Tables E.5 through E.8, E.13 through E.18)

TABLE 6. Relative Standard Deviations (RSDs) of Weight Gains for Nonirradiated UO_2 Fuel Pellets

Temperature, °C	Ref. Table	Number of Pellets	RSD, ^(a) %
150	E.2	18	(b)
180	E.6	5	13
200	E.8	8	25
230	E.16	8	25

(a) For 180, 200, and 230°C, the RSDs fluctuated over the given ranges during the first few weighing periods and then settled down to the relatively constant values.

(b) RSDs for 150°C ranged widely over all time periods, and did not settle down at longer times. This variance is due to the nearly zero total weight gain over much of the time at this temperature, coupled with the many small weight gains and losses observed.

At 150°C, neither theoretical density (TD) (or level of porosity) (100% TD = 10.96 g/cm³) nor surface area explained a significant portion of the pellet-to-pellet variation at any time. The surface condition was the same for all pellets at 150°C, and hence did not affect the results. At 180°C, TD had a statistically significant effect on weight percent gain at exposures of only 1133, 1270, and 1629 h with nonsignificant effects for all other times. Surface area had nonsignificant effects at all times at 180°C, and all pellets had the same surface condition. At 200°C, surface area had a statistically significant effect on weight gain only for times up to 454 h, with nonsignificant effects beyond 454 h. Theoretical density had a nonsignificant effect for all times, and all pellets had the same surface condition. Thus, for the most part, TD and surface area were not statistically significant in affecting pellet-to-pellet variability for tests on nonirradiated UO₂ between 150 and 200°C.

At 230°C, surface area, TD, and surface condition all had statistically significant effects on weight percent gain at 24 h. During the time period between 161 and 1042 h, only TD had a statistically significant effect.

Pellet aging (see next section) varied at the different temperatures but not within specimens at a given temperature, so it was not considered in the above analyses.

The failure of pellet density, surface area, and surface condition to have a statistically significant effect for the situations discussed above does not mean that their effects are never significant. The ranges of values for those variables (Tables E.1, E.5, E.7, and E.15) may have been too small to affect the results, or too small to allow declaration of the effects as statistically significant. Studies in which pellet characteristics varied over wider ranges are considered in the following subsections, but many of these studies did not incorporate the use of replicate samples, making statistical analysis of the data difficult or impossible.

Effect of Ambient Air Aging on UO₂ Oxidation

Ambient air aging (storage time in ambient air after hydrogen anneal and prior to testing) had a strong influence on the oxidation behavior. The

experimental results for pellets tested after different storage periods are presented in Appendix E. Increasing pellet aging 1) decreases the overall weight gain when U_3O_8 powder forms (Figure 25), 2) increases time to powder formation (Figure 26), and 3) decreases the initial rate of weight gain.

Differences in the oxidation behavior of UO_2 from different investigators (see Figures 27 through 30) may result in part from differences in pretest exposure times in air. Hastings and Novak (1983) initiated oxidation tests immediately after extracting fragments from CANDU fuel rods. Archive pellets tested by Tennessee Valley Authority (TVA) were stored in atmospheric air for several years prior to test initiation. Differences in oxidation behavior between CANDU and TVA pellets are consistent with the effect of ambient atmospheric aging. However, because microstructural features of TVA pellets were not identified,^(a) the possibility that the low oxidation rates in TVA pellets were due to microstructural features such as a large grain size cannot be ruled out.

FIGURE 25. Effect of Ambient Air Aging on Oxidation Behavior of Non-irradiated UO_2 Pellets at 200°C (Tables E.7 through E.12)

(a) NRC licensing agreements with TVA precluded transfer of nonirradiated pellets to PNL for evaluation.

FIGURE 26. Effect of Ambient Air Aging on the Time for Powder to Form in Nonirradiated UO_2 Pellets at 200°C (Tables E.7 through E.12)

FIGURE 27. Comparison of Oxidation Behavior of Nonirradiated UO_2 Pellets from Indicated Investigations at 250°C . White et al. (1983) data listed in Tables E.17 and E.18.

FIGURE 28. Comparison of Oxidation Behavior of Nonirradiated UO_2 Pellets at 230°C (Hastings and Novak 1983; White et al. 1983). White et al. (1983) data listed in Tables E.15 and E.16.

FIGURE 29. Comparison of PNL and TVA Data on Oxidation of Nonirradiated UO_2 Pellets at 200°C (PNL data listed in Tables E.9 and E.10)

FIGURE 30. Comparison of PNL and TVA Data on Oxidation of Nonirradiated UO_2 Pellets at 180°C (PNL data, listed in Tables E.5 and E.6, are from pellets that have aged 9 months.)

Aging time at ambient temperature reduced the rate of weight gain by oxidation and, presumably, extended the time for the onset of U_3O_8 powder formation. The magnitude of the effect on powder formation is shown in Figure 26. Two (White et al. 1983) of the specimens were examined by x-ray photoelectron spectroscopy (XPS) to determine differences in the chemical composition of the surfaces. The aged specimen was found to be coated with a large $\text{U}^{+6}/\text{U}^{+4}$ ratio, suggesting the presence UO_3 , which is consistent with the surface condition of UO_2 that was stored at ambient temperature for 15 years in air (Wadsten 1977). Colmenares (1975) has indicated a hydrated UO_3 surface film could provide a barrier to oxygen diffusion and thereby retard the rate of weight gain and delay the onset of U_3O_8 powder formation.

Effect of Surface Area on UO_2 Oxidation

Nonirradiated UO_2 pellets, which have a lower surface-area/volume ratio than spent fuel fragments, were mechanically fragmented to simulate an increased surface-area/volume ratio. The BET-measured surface area of the mechanically fragmented UO_2 was 35 times larger than the geometrical surface

area calculated for the UO_2 pellet. A comparison of the oxidation behavior of the nonirradiated UO_2 pellets and the mechanically fragmented pellets on the basis of weight percent is shown in Figure 31. The rate of weight gain prior to powder formation for the mechanically fragmented UO_2 was approximately five times higher than for the UO_2 pellets. A larger external surface area for the pellets (as determined by BET if it had been possible) would account for some of the differences in oxidation behavior between pellets and ground pellet fragments because the geometric surface area does not account for surface roughness. The time to powder formation as indicated in Figures 31 and 32 was not affected by the mechanical fragmentation of UO_2 pellets.

Weight gain from oxidation of specimens with different surface areas is commonly normalized to surface area, as is the case in Figure 32. As shown in Figure 32, this normalization procedure overcorrected for the increased surface area. Note that the weight per unit surface area parameter used in Figure 32

FIGURE 31. Effect of Fragmentation on Weight Gain for Nonirradiated UO_2 Pellets at 230°C (pellets aged 4 months in ambient air) (Tables I.1 and I.2)

FIGURE 32. Effect of Fragmentation on Weight Gain Normalized to Surface Area for Nonirradiated UO_2 Pellets at 230°C (pellets aged 4 months in ambient air) (Tables I.1 and I.2)

is not corrected for new surfaces created by the formation of U_3O_8 powder or for possible differences in surface roughness between pellets and mechanically fragmented UO_2 .

In a set of tests comparing oxidation rates of pellets from CRNL, UKCEGB-BNL, and PNL, the oxidation rate was found to vary inversely with the grain size as shown in Figure 33. This finding is consistent with the belief that grain boundaries provide the primary reaction surface (Simpson and Wood 1983; Einziger and Woodley 1985). Grains separate at their boundaries because of shrinkage associated with the formation of higher density U_4O_9 (Thomas, McCarthy, and Gilbert 1986). Later, the pressure resulting from the expansion of the U_3O_8 powder within grain boundaries causes further separation of grains. The expulsion of grains creates fresh UO_2 surfaces, as shown in Figure 34. The size of the powder particles formed by oxidation has been found to increase with increasing grain size (Kinugasa et al. 1982).

Surface area enhances UO_2 oxidation rates (Scott and Harrison 1963) by increasing the number of surface sites where oxygen enters the UO_2 lattice.

FIGURE 33. Initial Oxidation Rate Versus Grain Size for CRNL, UKCEGB-BNL, and PNL Nonirradiated UO_2 Pellets at 200 and 220°C (200 to 1600 h) (Tables J.1 to J.3)

FIGURE 34. Surface Spallation from U_3O_8 Formation in Nonirradiated UO_2 Pellet

Increased rates of weight gain in spent fuel, compared with nonirradiated pellets, have been at least partially attributed to the increased surface area from fragmentation of the fuel pellets (Hastings and Novak 1983; Simpson and Wood 1983). However, computations of the geometric surface area for PNL-generated data from nonirradiated pellets and irradiated fragments show that the difference in surface area can only partially account for the difference in weight gain kinetics. The remainder of the difference in kinetics must be explained by other factors, e.g., grain boundary surface area, fission product effects. PNL results are consistent with TEM (Thomas, McCarthy, and Gilbert 1986) and ceramographic studies (Wasywich et al. 1984; Einziger and Woodley 1985) of oxidized spent fuel that have shown the grain boundaries to be preferred paths for oxidation.

Effect of Pellet Density (Porosity) on UO_2 Oxidation

A comparison of weight gain data for nonirradiated UO_2 pellets in controlled-humidity air led to a correlation of results with pellet density (or porosity). The pellet densities, measured by levitation in distilled water, were found to vary randomly from 94 to 95.5% TD. The isochronal weight gain appeared to decrease with increasing TD (Figures 35 and 36) until a density of ~95% was achieved. A positive correlation of powder formation time with TD was found (Figure 37).

In the tests comparing oxidation of CRNL, UKCEGB-BNL, and PNL nonirradiated UO_2 pellets, the densities of the pellets ranged from 96.0 to 96.7% TD for PNL and CRNL pellets and 98.1 to 98.6% TD for the UKCEGB-BNL pellets. The weight gain versus time curves for the UKCEGB-BNL, PNL, and CRNL pellets are shown in Figures 38 and 39; the oxidation rate was higher for the lower-density PNL and CRNL pellets. At 220°C, U_3O_8 powder formed on freshly reduced CRNL and PNL pellets after 1200 h and on the higher-density UKCEGB-BNL pellets after 2000 to 2200 h (Figure 39). At 200°C, powder formed on the PNL pellets after 3000 h, and on the CRNL pellets after 3400 h, and no powder formed on the UKCEGB-BNL pellets after 3400 h (Figure 38). The time for U_3O_8 powder formation, therefore, was longest for the UKCEGB-BNL pellets and shortest for the PNL and CRNL pellets, indicating a positive correlation of powder formation time with density (porosity).

FIGURE 35. Effect of Density on Weight Gain for Nonirradiated UO_2 Pellets After 363 h at 220°C (Tables G.3 and G.4)

FIGURE 36. Effect of Density on Weight Gain for Nonirradiated UO_2 Pellets After 756 h at 220°C (Tables G.3 and G.4)

FIGURE 37. Effect of Density on the Time to Powder Formation for Nonirradiated UO_2 Pellets at 220°C (Tables G.3 and G.4)

FIGURE 38. Weight Gain Versus Time for CRNL, UKCEGB-BNL, and PNL Nonirradiated UO_2 Pellets at 200°C (Tables J.1 and J.2, freshly recuded CRNL, UKCEGB-BNL, and PNL pellets plotted)

FIGURE 39. Weight Gain Versus Time for CRNL, UKCEGB-BNL, and PNL Nonirradiated UO_2 Pellets at 220°C (Tables J.1 and J.3, freshly reduced CRNL, UKCEGB-BNL, and PNL pellets plotted)

The effect of UO_2 density on the time for U_3O_8 powder formation was identified by Simpson and Wood (1983) by comparing the oxidation behavior of UO_2 specimens with different densities. In their study at 250°C , the induction time to U_3O_8 powder formation varied from 60 h for a density of 94.5% TD to 345 h for a density of 98.5% TD. The rates of weight gain for the fuels with different densities were similar.

The variation of the time to U_3O_8 powder formation with density as shown in Figure 39 is consistent with the trend identified by Simpson and Wood (1983). If this trend is interpreted to result from a greater number of surface reaction sites associated with a decrease in density, it seems inconsistent with the insensitivity of powder formation time to surface area shown in Figures 31 and 32. The insensitivity of the oxidation rate to densities above 94.5% (Figures 35 and 36) also seems to be inconsistent with surface area effects. These findings are consistent if there is limited open porosity at densities greater than 94.5%. Therefore, density increases above 94.5% may not

decrease the number of reaction sites for UO_2 oxidation. Below 94.5% TD, oxidation kinetics increase with decreasing density. The densities of the pellets tested by Simpson and Wood (1983) were in the range of 94.5 to 98.6% TD and, therefore, may have had limited open porosity.

The results appear to be consistent with data on single-crystal UO_2 (Wang 1981), which was more resistant to powder formation than polycrystalline UO_2 . The high-density UO_2 pellets (Simpson and Wood 1983) appear to approach the oxidation behavior of single-crystal UO_2 . Apparently, the crystalline imperfections at the grain boundaries are less severe in the high-density specimens, thereby reducing the short-circuited diffusion of oxygen along grain boundaries.

Effect of Moisture on UO_2 Oxidation

A literature review indicated that moisture was not an important oxidation parameter (Bannister 1968). Einziger and Woodley (1985) found moisture had little effect on spent fuel oxidation. However, tests at PNL showed that moisture had some effect over the range of 0 (<5 ppm) to 160 mg/L (60°C dew point) for oxidation of nonirradiated UO_2 pellets at 230 and 200°C. The initial oxidation rate for nonirradiated pellets at 200 and 230°C increased with increasing moisture (Figures 40 and 41). A factor of approximately 4 increase in weight gain with increasing moisture was found at various times over the range of moisture in these tests (Figures 42 and 43). Moisture levels of 20 and -7°C dew point resulted in initially higher oxidation rates but longer powder formation periods than for a moisture level of -40°C dew point (Tables G.1 and G.2).

The PNL pellet results (Appendix G) are qualitatively consistent with the effects of moisture on powder at 113°C (National Bureau of Standards 1956), as shown in Figure 44. The reactivity of the high surface area of the powder enabled measurement of weight gains at considerably lower temperatures than possible in pellets. High-surface-area powders may also have made the powder more susceptible to moisture effects on the oxidation rate.

Attempts to develop a data base (Appendix G) over a narrower range of moisture levels (-35 to 20°C dew point where an increase of a factor of 2 in

FIGURE 40. Effect of Moisture Levels in Air on Oxidation of Nonirradiated UO_2 Pellets at 200°C (Tables G.5 and G.6)

FIGURE 41. Effect of Moisture Levels in Air on Oxidation of Nonirradiated UO_2 Pellets at 230°C (Tables G.5 and G.6)

FIGURE 42. Effect of Moisture on Oxidation of Nonirradiated UO_2 Pellets at 230°C and 377 h (Tables G.5 and G.6)

FIGURE 43. Effect of Moisture on Oxidation of Nonirradiated UO_2 Pellets at 200°C and 1419 h (Tables G.5 and G.6)

weight gain was predicted from the data in Figures 42 and 43) were masked by the effect of variation in pellet density.

Effect of Thermal Cycling on UO_2 Oxidation

During the tests, the UO_2 pellets were periodically cooled to perform visual examinations and intermittent weighings, resulting in thermal cycling of the pellets. The oxidation behavior of pellets with different thermal cycling histories was compared. Two pellets were weighed five times over a 3911-h time period. One pellet was thermally cycled three times between interruptive

FIGURE 44. Effect of Moisture on Oxidation of UO_2 at 113°C
(National Bureau of Standards 1956)

weighing until it was exposed to a total of 18 cycles between ambient temperature and the 200°C test temperature. The weight gained at powder formation by the pellet with 18 cycles was similar to that gained by the pellet with only five cycles. The average rate of weight increase for the two pellets was $0.0014 \text{ mg/cm}^2/\text{h}$. The initiation of powder spallation in these pellets, which were aged 8 months at ambient temperature after being hydrogen annealed, occurred after 2800 h for both thermal cycling conditions. These results are plotted as weight increase versus time in Figure 45. The results shown in Figure 45 indicate that there is no significant effect due to thermal cycling between the 200°C test temperature and the ambient weighing temperature. The heating and cooling rates are too slow to induce significant temperature gradients and stresses across spent fuel fragments that could cause cracking and induce new surface formation. Therefore, test procedures utilizing pellet cooling for weight measurement and those using isothermal continuous weighing are essentially equivalent. Further confirmation of the effects of thermal cycling on oxidation behavior may be required for spent fuel.

FIGURE 45. Effect of Thermal Cycles Between Ambient Temperature and 200°C Test Temperature for Nonirradiated UO_2 Pellets Aged 8 Months in Ambient Air Before Testing

Effect of Test-Temperature Change on UO_2 Oxidation

Test-temperature changes were performed to provide data for damage models that predict oxidation behavior under changing temperature conditions. Tests were initiated in air with UO_2 pellets at 150°C, 200°C, and 230°C. At a specified fraction of the powder formation period, selected specimens were transferred to a furnace at a different temperature. Some pellets were allowed to continue without a change in temperature. The results are shown in Figures 46 and 47 for pellets aged 4 months (150°C data in Table K.6 not shown in Figure 46). The weight gain and the powder formation period were affected by the combined time at 230°C and 200°C. A comprehensive analysis of these data has been published (Reid and Gilbert 1986).

Effect of Oxidants on UO_2 Oxidation

The effect of N_xO_y oxidants on the stability of UO_2 is of interest because N_xO_y compounds form by radiolysis during irradiation of air in a gamma radiation field. Up to 0.25 vol% N_2O was found in analysis of an air cover gas surrounding a PWR spent fuel assembly (Johnson et al. 1985).

FIGURE 46. Effect of Temperature Change from 200 to 230°C on Oxidation Behavior of Nonirradiated UO_2 Pellets Aged 4 Months in Ambient Air (Tables K.1 through K.4, I.1, and I.2)

FIGURE 47. Effect of Temperature Change from 230 to 200°C on Oxidation Behavior of Nonirradiated UO_2 Pellets Aged 4 Months in Ambient Air (Tables K.1, K.2, and K.5)

Air containing 1 vol% NO_2 was found to accelerate the rate of weight gain significantly and to reduce the time to powder formation (Figures 48 and 49). The weight gain for UO_3 formation exceeded the 4% (Figure 48) associated with stoichiometric conversion of UO_2 to U_3O_8 powder. The oxidation led to the formation of UO_3 and not U_3O_8 that is usually found in tests in air without the presence of oxidants. The third pellet tested in the presence of NO_2 at 215°C (triangular symbols in Figure 49) had a 1% lower TD value than the other two pellets (95% versus 96%). The higher rate of oxidation is attributed to lower density. A concentration of 1% NO_2 may not be typical of the levels formed through radiolysis in a dry storage atmosphere, but the qualitative effect of a strong oxidant such as NO_2 on UO_2 oxidation is clear.

Gamma irradiation from a cobalt-60 source increased the oxidation rate of nonirradiated UO_2 pellets. Results at 200, 210, and 220°C after 200 h are shown in Figure 50 for pellets heated in hermetically sealed ovens (static cover gas). No gamma irradiation effects were found in tests conducted in a

FIGURE 48. Nonirradiated UO_2 Pellet Oxidation in Air and Air-1 vol% NO_2 at 250°C Aged 1 Month in Ambient Air (Tables F.3 and F.4)

FIGURE 49. Nonirradiated UO_2 Pellet Oxidation in Air and Air-1 vol% NO_2 at 215°C Aged 4 Months in Ambient Air (Tables F.1 and F.2)

FIGURE 50. Effect of Gamma Radiation on Oxidation of Nonirradiated UO_2 (additional data are listed in Tables H.1 to H.3)

cover gas that was connected to a flowing gas source. Apparently, sufficient concentrations of N_xO_y oxidant compounds will accumulate only in the cover gas of a sealed system. In a system where the cover gas is constantly being exchanged, the radiolysis products become diluted by flushing of the cover gas as air is replenished.

The suspected oxidant is NO_2 because NO_2 was noticeably missing in the cover gas analyses but was found to be very reactive during UO_2 oxidation tests in a mixture of air with 1 vol% NO_2 (Figures 48 and 49). Anderson et al. (1955) reported enhanced UO_2 oxidation in the presence of NO_2 .

Effect of Gadolinia Additions on UO_2 Oxidation

Gadolinia (Gd_2O_3) is used as an additive to UO_2 in nuclear reactor fuel. Gadolinium is a "burnable poison" that has two high-neutron adsorption cross section isotopes (^{155}Gd and ^{151}Gd) that transmute to low cross section isotopes (^{156}Gd and ^{158}Gd) after several months of reactor operation. Gadolinia is accommodated in the UO_2 crystal lattice by the formation of U^{+6} or vacant oxygen sites to maintain electrical neutrality (Grossman, Packard, and Hill 1972; Ohmichi et al. 1981; Une and Oguma 1983; Catlow 1979).

A faster rate of weight gain was found for UO_2 pellets containing gadolinia than for nongadolinia-bearing pellets. As shown in Figure 51, gadolinia increased the weight gain at which powder formation occurred and the time to powder formation. Powders produced by oxidation of 5 wt% and 10 wt% (7.27 at.% and 14.16 at.%, respectively) gadolinia were examined by XRD. At high diffraction angles the pattern split, indicating the presence of an oxide with the tetragonal symmetry of U_3O_7 . The spalled material also produced weak diffraction peaks that are characteristic of alpha U_3O_8 or UO_3 . Strong sharp peaks were also found that correspond to face-centered-cubic UO_2 with gadolinia in solution. The lattice parameter in both oxidized 5 wt% and 10 wt% gadolinia UO_2 pellets was 5.425 Å. This was significantly less than the 5.469 Å for nonoxidized UO_2 , 5.460 Å for nonoxidized UO_2 containing 5 wt% gadolinia, and 5.466 Å for nonoxidized UO_2 containing 10 wt% gadolinia.

FIGURE 51. Effect of Gadolinia on Oxidation of Nonirradiated UO₂ Pellets at 200°C Aged 22 Months in Ambient Air (Tables E.9 and E.10, pellets E1, 5E1, and 10E1)

The UO₂ gadolinia pellets broke into many fragments having a typical dimension of about 1 mm shortly after U₃O₈ formed. UO₂ pellets usually form powder with dimensions less than 10 μm from expulsion of single grains prior to fragmentation (White et al. 1983).

The reduction of the lattice parameter from 5.460 Å for nonoxidized UO₂ containing 5 wt% gadolinia and from 5.466 Å for nonoxidized UO₂ containing 10 wt% gadolinia to 5.424 Å in the powder formed from oxidizing the UO₂-gadolinia compositions is consistent with the effect of filling vacant oxygen lattice sites with oxygen. The attractive forces between the oxygen valence electrons and the lattice contract the UO₂ lattice and reduce the lattice parameter.

FIGURE 81. Effect of gadolinia on oxidation of nonirradiated UO₂ pellets at 200°C aged 22 months in ambient air. (Tables E.9 and E.10, pellets E1, E2, and E3)

The UO₂ gadolinia pellets broke into many fragments having a typical dimension of about 1 mm shortly after U₃O₈ formed. UO₂ pellets usually form powder with dimensions less than 10 µm from expansion of single grains prior to fragmentation (White et al., 1983).

The reduction of the lattice parameter from 2.450 Å for nonoxidized UO₂ containing 5 wt% gadolinia and from 2.466 Å for nonoxidized UO₂ containing 10 wt% gadolinia to 2.454 Å in the powder formed from oxidizing the UO₂-gadolinia compositions is consistent with the effect of filling vacant oxygen lattice sites with oxygen. The attractive forces between the oxygen valence electrons and the lattice contract the UO₂ lattice and reduce the lattice parameter.

CONCLUSIONS

1. Oxidation behavior of spent fuel differs markedly from nonirradiated UO_2 pellets. Initial rates of weight gain of PWR spent fuel are 40 to 60 times higher, the period for powder to form is longer, and powder formation begins at higher levels of weight gain.
2. Oxidation tests on different PWR-type spent fuels produced a general increase in weight gain with both increasing temperature (135 to 200°C) and increasing exposure time up to 5100 h. PWR fuel type and burnup had a significant influence on weight gain. The rate of weight gain also appeared to increase with microporosity.
3. Comparison of the oxidation behavior to the microstructures of three different types of nonirradiated pellets showed that the oxidation rate increased with decreasing grain size.
4. The period for U_3O_8 powder to form on nonirradiated UO_2 is independent of geometric surface area, and increases with increasing density and increasing exposure period to ambient air prior to elevated temperature testing.
5. Application of a gamma field of 10^5 R/h significantly increases the rate of weight gain of nonirradiated UO_2 pellets under a sealed cover gas system, presumably resulting from the buildup of concentrations of gaseous products of radiolysis that are strong oxidants. Gamma radiation appears to have less of an effect, if any, on spent fuel weight gain than on nonirradiated UO_2 pellets.
6. Increasing amounts of moisture (5 ppm to 60°C dew point) in air increased the initial rate of oxidation of nonirradiated UO_2 pellets. Preliminary results on spent PWR fuel indicate that moisture (0 to 40°C dew point) does not significantly influence the rate of oxidation.

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APPENDIX A

SPENT FUEL TESTS IN AIR (POINT BEACH FUEL)

TABLE A.1. Initial Characteristics of Fuel Samples for 200°C Oxidation Tests on Irradiated UO₂ (Spent Fuel) in Air in Ventilated Ovens

Sample Number	Fuel Type	Sample Location, in. ^(a)	Average Burnup, GWd/MTU	Gamma Field, R/h	Weight, g	Reactor Discharge Date
1E	Point Beach (fragments)	49.25 - 47.25	29.5	600	39.184	May 1974
1F	NI ^(b)	N/A ^(c)	N/A	600	10.649	N/A
1G	Point Beach (fragments)	53.25 - 51.25	29.5	600	33.411	May 1974
1H	Point Beach (rod segments)	40.38 ^(b)	29.5	600	44.542	May 1974
2E	Point Beach	41.25 - 39.25	29.5	150,000	37.342	May 1974
2F	NI	N/A	N/A	150,000	10.797	N/A
2G	Point Beach	51.25 - 49.25	29.5	150,000	34.207	May 1974
2H	Point Beach	40.38 ^(d)	29.5	150,000	35.325	May 1974

(a) Referenced to sealed end of rod PB-A.

(b) NI = Nonirradiated pellets.

(c) N/A = Not applicable.

(d) Average sample location.

TABLE A.2. Weight Gain (wt%) for 200°C Oxidation Tests on Irradiated UO_2 in Air (600 R/h) in Ventilated Ovens

Time at Temperature, h	Sample Number			
	1E	1F	1G	1H
0	0	0	0	0
65.33	-0.003	-0.047	0.003	0.022
185.33	0.082	-0.056	0.096	-0.020
344.75	0.212	-0.056	0.293	0.155
402.17	0.258	-0.038	(a)	0.213
540.25	0.347	-0.009		0.346
750.50	0.470	-0.047		0.485
881.25	0.554 ^(b)	0.019		0.566
1063.92	0.661	-0.122		0.707
1254.42	0.773	-0.019		0.831
1421.59	0.845	0.019		0.918
1561.22	0.845	0.019		0.918
1754.63	0.845	0.019		0.918
2015.71	1.305	0.649		1.168
2251.56	1.415	0.639		1.278
2762.64	1.645	0.759		1.508
2941.47	1.725 ^(c)	0.769		1.598 ^(c)
5531.47	1.915	0.789		1.828

(a) Sample removed from test for cumulative damage studies, sample split; part returned to 200°C test, remainder of sample placed in 230°C test.

(b) Corrected for 0.398 g of fuel spillage.

(c) Fine particulates approx. 1 mm on edge detected.

TABLE A.3. Weight Gain (wt%) for 200°C Oxidation Tests on Irradiated UO_2 in Air (150,000 R/h) in Ventilated Ovens

Time at Temperature, h	Sample Number			
	2E	2F	2G	2H
0	0	0	0	0
65.33	0.396	0.028	0.009	-0.023
185.33	0.485	-0.028	0.082	(a)
(b)				

(a) Most of sample was spilled.

(b) Oven failure - test terminated.

TABLE A.4. Initial Characteristics of Fuel Samples for 230°C Oxidation on Irradiated UO₂ (Spent Fuel) in Air in Ventilated Ovens

Sample Number	Fuel Type	Sample Location, in. ^(a)	Average Burnup, GWd/MTU	Gamma Field, R/h	Weight, g
1A	Point Beach (rod segments)	64.25 - 63.25 60.25 - 59.25 ^(b)	29.5	600	35.941
1B	NI ^(c)	N/A ^(d)	N/A	600	15.328
1C	Point Beach	65.25 - 64.25 63.25 - 62.25 ^(b)	29.5	600	34.181
1D	Point Beach (fragments)	62.25 - 60.25	29.5	600	30.884
2A	Point Beach (rod segments)	55.25 - 53.25	29.5	150,000	31.703
2B	NI	N/A	N/A	150,000	16.007
2C	Point Beach	59.25 - 57.25	29.5	150,000	31.546
2D	Point Beach (fragments)	57.25 - 55.25	29.5	150,000	33.173

(a) Referenced to sealed end of rod PB-A.

(b) Samples taken from two rod sections.

(c) NI = Nonirradiated pellets.

(d) N/A = Not applicable.

TABLE A.5. Weight Gain (wt%) for 230°C Oxidation Tests on Irradiated UO₂ in Air (600 R/h) in Ventilated Ovens

Time at Temperature, h	Sample Number ^(a)			
	1A	1B	1C	1D
0.0	0.0	0.0	0.0	0.0
15.58	0.0	-0.01	-4.10	0.18
33.11	0.05	-0.01	-4.02	0.28
54.86	0.14	-0.01	(b)	0.41
78.69	0.24	-0.01		0.70
101.27	0.34	-0.05		0.93
150.10	0.51	-0.01		1.10
168.90	0.58	-0.02		1.21
188.48	0.66	-0.02		(c)
210.81	0.72	-0.03		1.68
260.19	0.85	0.0		2.00
283.19	0.93	-0.01		2.11
308.52	1.03	-0.01		2.21
325.97	1.08	-0.01		2.29
396.89	1.27	0.0		2.50
437.97	1.36	0.02		2.60
485.80	1.46	0.05		2.71
557.54	1.57	0.05		2.83
619.39	1.62	0.03		2.87
672.30	1.69	0.05		2.93
764.88	1.79	0.07		3.01
835.96	1.84	0.07		3.04
996.43	(d)	0.07 ^(e)		3.04
1190.85		0.07		3.04
1451.68		0.37		3.23
1687.09		0.41		3.23
2198.26		0.67		3.24
2834.94		1.09		3.25 ^(f)
5427.86		2.98		3.27

(a) Sample weight gains corrected for fuel spillage with the exception of 1C.

(b) Sample removed from test.

(c) Inconsistent data, probably due to weighing error.

(d) Sample moved to 200°C oven.

(e) U₃O₈ powder formation on nonirradiated pellets.

(f) Fine particulates approx. 1 mm on edge detected.

TABLE A.6. Weight Gain (wt%) for 230°C Oxidation Tests on Irradiated UO_2 in Air (150,000 R/h) in Ventilated Ovens

Time at temperature, h	Sample Number ^(a)			
	2A	2B	2C	2D
0.0	0.0	0.0	0.0	0.0
18.75	0.01	-0.01	0.05	0.06
39.75	0.08	0.01	0.20	0.21
63.33	0.18	0.0	(b)	0.39
85.41	0.26	0.01		0.55
133.91	0.44	0.03		0.69
152.57	0.49	0.06		0.78
171.99	0.5	0.02		0.90
194.24	0.56	0.02		1.00
243.53	0.71	0.04		1.19
266.78	0.73	0.04		1.28
292.03	0.80	0.03		1.37
314.36	0.87	0.05		1.44
384.61	0.95	0.06		1.57
430.52	1.06	0.06		1.66
478.27	1.16	0.07		1.75
549.94	1.29	0.08 ^(c)		1.87
611.62	1.33	0.11		1.95
664.12	1.41	0.11		1.99
756.70	1.49	0.12		2.05
827.53	1.54	0.15		2.08
1126.26	1.54	0.15		2.08
1320.68	1.54	0.15		2.08
1581.51	1.84	0.52		2.34
1816.92	1.83	0.61		2.33
2328.09	1.88	1.16		2.39
2826.51	1.91	2.01		2.40
5419.43	2.02	4.12		2.46

(a) Sample weight gains corrected for fuel spillage with the exception of 2C.

(b) Sample removed from test.

(c) U_3O_8 powder formation on nonirradiated pellets.

APPENDIX B

CONTROLLED-ATMOSPHERE SPENT FUEL TESTS
IN LOW GAMMA FIELD

TABLE B.1. Low-Gamma Air Oxidation Test Matrix Actual Operating Temperatures of Hermetically Sealed Ovens^(a)

Oven #	Nominal Temp.	Average Shelf Temperature, °C ^(b)		
		Top	Middle	Bottom
1	135	132 (±4)	125 (±4)	N/A
2	200	203 (±2)	198 (±3)	N/A
3	150	150 (±1)	143 (±2)	N/A
4	180	185 (±1)	175 (±1)	N/A
6	165	172 (±1)	162 (±3)	N/A

(a) Samples Y-1 through Y-4 (where Y = oven number) located on top oven shelf. Samples Y-5 through Y-6 located on middle oven shelf. Bottom shelf was not used.

(b) Standard deviation in parenthesis. Averages determined from daily measurements.

TABLE B.2. Cover Gas Analyses for Low-Gamma-Field Oxidation Tests in Air on Spent PWR Fuel in Hermetically Sealed Ovens^(a)

Oven #	Run # ^(b)	Concentration, mol%							
		CO ₂	Ar	O ₂	N ₂	CO	He	H ₂	CH ₄
1	1	0.14	0.83	21.1	77.8	<0.1	0.06	<0.01	<0.01
1	2	0.41	0.33	19.7	78.0	<0.1	1.56	<0.01	<0.01
2	1	0.87	0.17	13.0	85.2	<0.1	0.74	<0.01	<0.01
2	2	0.41	0.23	18.9	80.1	<0.1	0.35	<0.01	<0.01
3	1	0.59	0.15	19.8	79.2	<0.1	0.28	<0.01	<0.01
3	2	1.12	0.18	19.8	78.4	<0.1	0.44	<0.01	<0.01
4	1	0.64	0.16	17.3	81.7	<0.1	0.20	<0.01	<0.01
4	2	0.63	0.21	18.7	80.1	<0.1	0.29	<0.01	<0.01
6	1	0.34	0.22	18.0	80.6	<0.1	0.86	<0.01	<0.01
6	2	1.08	0.29	18.2	79.4	<0.1	1.05	<0.01	<0.01

(a) Kr and Xe not analyzed for.

(b) A run is defined as a period of time at test temperature prior to an interim weighing.

TABLE B.3. Results of Tests with Spent PWR Fuel in Air at 135°C
(500 to 1000 R/h) in a Hermetically Sealed Oven

Specimen	Fuel Type ^(a) (Section)	Burnup, ^(b) GWd/MTU	Dew Point, °C	Time, h	Weight, g	Wt. Gain, wt%
1-1	NI	0	27.9	0	15.949	0
1-2	HBR (2I)	30.9	27.9	0	30.239	0
1-3	PB "A"	29.5	27.9	0	32.018	0
1-4	PB "A"	LBU	27.9	0	26.505	0
1-5	SP "B"	23.7	27.9	0	27.662	0
1-1	NI	0	27.9	1537.3	15.952	0.019
1-2	HBR (2I)	30.9	27.9	1537.3	30.241	0.007
1-3	PB "A"	29.5	27.9	1537.3	32.022	0.012
1-4	PB "A"	LBU	27.9	1537.3	26.508	0.011
1-5	SP "B"	23.7	27.9	1537.3	27.663	0.004
1-1	NI	0	30.9	1537.3	15.952	(a)
1-2	HBR (2I)	30.9	30.9	1537.3	30.283	(a)
1-3	PB "A"	29.5	30.9	1537.3	32.015	(a)
1-4	PB "A"	LBU	30.9	1537.3	26.501	(a)
1-5	SP "B"	23.7	30.9	1537.3	27.659	(a)
1-1	NI	0	30.9	4727.9	15.959	0.063
1-2	HBR (2I)	30.9	30.9	4727.9	30.283	0.007
1-3	PB "A"	29.5	30.9	4727.9	32.024	0.041
1-4	PB "A"	LBU	30.9	4727.9	26.507	0.034
1-5	SP "B"	23.7	30.9	4727.9	27.685	0.098

(a) NI = Nonirradiated fuel pellets

HBR = H. B. Robinson (Section)

PB = Point Beach ("A" rod)

SP = Shippingport ("B" rod)

(b) Burnups for Point Beach (PB) and Shippingport (SP) are averages.

Burnups for H. B. Robinson (HBR) determined from burnup analysis of rod and gamma scan plots (ref.: Barner 1985, PNL-5109 Rev. 1).

LBU = Low burnup samples sectioned from low power end of rod.

Estimated burnup = 20 GWd/MTU.

(c) Starting conditions for second interim weighing period (Run 2).

Sample weigh boats changed between end of first weighing period and beginning of second weighing period.

TABLE B.4. Results of Tests with Spent PWR Fuel in Air at 150°C
(500 to 1000 R/h) in a Hermetically Sealed Oven

Specimen	Fuel Type ^(a) (Section)	Burnup, ^(b) GWd/MTU	Dew Point, °C	Time, h	Weight, g	Wt. Gain, wt%
3-1	NI	0	27.9	0	15.946	0
3-2	HBR (1I)	30.9	27.9	0	29.210	0
3-3	HBR (1J)	22.9	27.9	0	33.299	0
3-4	PB "A"	29.5	27.9	0	32.550	0
3-5	PB "A"	LBU	27.9	0	23.781	0
3-6	SP "B"	23.7	27.9	0	23.074	0
3-1	NI	0	27.9	1518.8	15.950	0.025
3-2	HBR (1I)	30.9	27.9	1518.8	29.232	0.075
3-3	HBR (1J)	22.9	27.9	1518.8	33.308	0.027
3-4	PB "A"	29.5	27.9	1518.8	35.574	0.074
3-5	PB "A"	LBU	27.9	1518.8	23.783	0.008
3-6	SP "B"	23.7	27.9	1518.8	23.195	0.524
3-1	NI	0	29.6	1518.8	15.948	(c)
3-2	HBR (1I)	30.9	29.6	1518.8	29.233	(c)
3-3	HBR (1J)	22.9	29.6	1518.8	33.304	(c)
3-4	PB "A"	29.5	29.6	1518.8	32.567	(c)
3-5	PB "A"	LBU	29.6	1518.8	23.779	(c)
3-6	SP "B"	23.7	29.6	1518.8	23.191	(c)
3-1	NI	0	29.6	4886.8	15.958	0.088
3-2	HBR (1I)	30.9	29.6	4886.8	29.690	1.639
3-3	HBR (1J)	22.9	29.6	4886.8	33.343	0.144
3-4	PB "A"	29.5	29.6	4886.8	32.641	0.301
3-5	PB "A"	LBU	29.6	4886.8	23.796	0.080
3-6	SP "B"	23.7	29.6	4886.8	23.279	0.906

(a) NI = Nonirradiated fuel pellets

HBR = H. B. Robinson (Section)

PB = Point Beach ("A" rod)

SP = Shippingport ("B" rod)

(b) Burnups for Point Beach (PB) and Shippingport (SP) are averages.

Burnups for H. B. Robinson (HBR) determined from burnup analysis of rod and gamma scan plots (ref.: Barner 1985, PNL-5109 Rev. 1).

LBU = Low burnup samples sectioned from low power end of rod.

Estimated burnup = 20 GWd/MTU.

(c) Starting conditions for second interim weighing period (Run 2).

Sample weigh boats changed between end of first weighing period and beginning of second weighing period.

TABLE B.5. Results of Tests with Spent PWR Fuel in Air at 165°C
(500 to 1000 R/h) in a Hermetically Sealed Oven

Specimen	Fuel Type ^(a) (Section)	Burnup, ^(b) GWd/MTU	Dew Point, °C	Time, h	Weight, g	Wt. Gain, wt%
6-1	NI	0	27.9	0	15.690	0
6-2	HBR (1Q)	30.9	27.9	0	14.622	0
6-3	PB "A"	29.5	27.9	0	30.420	0
6-4	PB "A"	LBU	27.9	0	28.610	0
6-5	SP "B"	23.7	27.9	0	27.953	0
6-1	NI	0	27.9	1539.2	15.692	0.013
6-2	HBR (1Q)	30.9	27.9	1539.2	14.752	0.889
6-3	PB "A"	29.5	27.9	1539.2	30.615	0.641
6-4	PB "A"	LBU	27.9	1539.2	28.684	0.259
6-5	SP "B"	23.7	27.9	1539.2	28.435	1.724
6-1	NI	0	30.1	1539.2	15.694	(c)
6-2	HBR (1Q)	30.9	30.1	1539.2	14.752	(c)
6-3	PB "A"	29.5	30.1	1539.2	30.607	(c)
6-4	PB "A"	LBU	30.1	1539.2	28.682	(c)
6-5	SP "B"	23.7	30.1	1539.2	28.430	(c)
6-1	NI	0	30.1	5040.2	15.704	0.076
6-2	HBR (1Q)	30.9	30.1	5040.2	14.901	1.908
6-3	PB "A"	29.5	30.1	5040.2	30.865	1.489
6-4	PB "A"	LBU	30.1	5040.2	28.902	1.028
6-5	SP "B"	23.7	30.1	5040.2	28.592	2.304

- (a) NI = Nonirradiated fuel pellets
HBR = H. B. Robinson (Section)
PB = Point Beach ("A" rod)
SP = Shippingport ("B" rod)
- (b) Burnups for Point Beach (PB) and Shippingport (SP) are averages.
Burnups for H. B. Robinson (HBR) determined from burnup analysis of rod and gamma scan plots (ref.: Barner 1985, PNL 5109 Rev. 1).
LBU = Low burnup samples sectioned from low power end of rod.
Estimated burnup = 20 GWd/MTU.
- (c) Starting conditions for second interim weighing period (Run 2).
Sample weigh boats changed between end of first weighing period and beginning of second weighing period.

TABLE B.6. Results of Tests with Spent PWR Fuel in Air at 180°C
(500 to 1000 R/h) in a Hermetically Sealed Oven

Specimen	Fuel Type ^(a) (Section)	Burnup, ^(b) Gwd/MTU	Dew Point, °C	Time, h	Weight, g	Wt. Gain, wt%
4-1	NI	0	28.2	0	15.915	0
4-2	HBR (1Q)	30.9	28.2	0	32.410	0
4-3	HBR (2J)	20.9	28.2	0	27.619	0
4-4	PB "A"	29.5	28.2	0	30.814	0
4-5	PB "A"	LBU	28.2	0	26.029	0
4-6	SP "A"	23.7	28.2	0	36.261	0
4-1	NI	0	28.2	1705.75	15.912	-0.019
4-2	HBR (1Q)	30.9	28.2	1705.75	32.606	0.605
4-3	HBR (2J)	20.9	28.2	1705.75	27.736	0.424
4-4	PB "A"	29.5	28.2	1705.75	30.982	0.545
4-5	PB "A"	LBU	28.2	1705.75	26.067	0.146
4-6	SP "A"	23.7	28.2	1705.75	36.826	1.558
4-1	NI	0	30.7	1705.75	15.917	(c)
4-2	HBR (1Q)	30.9	30.7	1705.75	32.602	(c)
4-3	HBR (2J)	20.9	30.7	1705.75	27.735	(c)
4-4	PB "A"	29.5	30.7	1705.75	30.978	(c)
4-5	PB "A"	LBU	30.7	1705.75	26.061	(c)
4-6	SP "A"	23.7	30.7	1705.75	36.161	(c)
4-1	NI	0	30.7	5113.03	15.931	0.069
4-2	HBR (1Q)	30.9	30.7	5113.03	32.915	1.571
4-3	HBR (2J)	20.9	30.7	5113.03	27.961	1.242
4-4	PB "A"	29.5	30.7	5113.03	31.241	1.399
4-5	PB "A"	LBU	30.7	5113.03	25.715 ^(d)	-1.183
4-6	SP "A"	23.7	30.7	5113.03	36.361	2.110

(a) NI = Nonirradiated fuel pellets

HBR = H. B. Robinson (Section)

PB = Point Beach ("A" rod)

SP = Shippingport ("A" rod)

(b) Burnups for Point Beach (PB) and Shippingport (SP) are averages.

Burnups for H. B. Robinson (HBR) determined from burnup analysis of rod and gamma scan plots (ref.: Barner 1985, PNL 5109 Rev. 1).

LBU = Low burnup samples sectioned from low power end of rod.

Estimated burnup = 20 Gwd/MTU.

(c) Starting conditions for second interim weighing period (Run 2).

Sample weigh boats changed between end of first weighing period and beginning of second weighing period.

(d) Erratic data point, fuel loss suspected.

TABLE B.7. Results of Tests with Spent PWR Fuel in Air at 200°C
(500 to 1000 R/h) in a Hermetically Sealed Oven

Specimen	Fuel Type ^(a) (Section)	Burnup, ^(b) GWd/MTU	Dew Point, °C	Time, h	Weight, g	Wt. Gain, wt%
2-1	NI	0	27.5	0	16.148	0
2-2	HBR (1Q)	30.9	27.5	0	32.234	0
2-3	HBR (T)	22	27.5	0	52.285	0
2-4	PB "A"	29.5	27.5	0	31.067	0
2-5	PB "A"	LBU	27.5	0	16.081	0
2-6	SP "A"	23.7	27.5	0	37.478	0
2-1	NI	0	27.5	1679.0	16.160	0.074
2-2	HBR (1Q)	30.9	27.5	1679.0	32.734	1.551
2-3	HBR (T)	22	27.5	1679.0	53.169	1.691
2-4	PB "A"	29.5	27.5	1679.0	31.451	1.236
2-5	PB "A"	LBU	27.5	1679.0	16.159	0.485
2-6	SP "A"	23.7	27.5	1679.0	38.239	2.031
2-1	NI	0	29.6	1679.0	16.168	(c)
2-2	HBR (1Q)	30.9	29.6	1679.0	32.736	(c)
2-3	HBR (T)	22	29.6	1679.0	53.161	(c)
2-4	PB "A"	29.5	29.6	1679.0	32.450	(c)
2-5	PB "A"	LBU	29.6	1679.0	15.723	(c)
2-6	SP "A"	23.7	29.6	1679.0	38.236	(c)
2-1	NI	0	29.6	5069.5	16.240	0.520
2-2	HBR (1Q)	30.9	29.6	5069.5	32.992	2.345
2-3	HBR (T)	22	29.6	5069.5	53.469	2.280
2-4	PB "A"	29.5	29.6	5069.5	31.779	2.295
2-5	PB "A"	LBU	29.6	5069.5	15.984	2.108
2-6	SP "A"	23.7	29.6	5069.5	38.333	2.289

- (a) NI = Nonirradiated fuel pellets
HBR = H. B. Robinson (Section)
PB = Point Beach ("A" rod)
SP = Shippingport ("A" rod)
- (b) Burnups for Point Beach (PB) and Shippingport (SP) are averages.
Burnups for H. B. Robinson (HBR) determined from burnup analysis of rod and gamma scan plots (ref.: Barner 1985, PNL-5109 Rev. 1).
LBU = Low burnup samples sectioned from low power end of rod.
Estimated burnup = 20 GWd/MTU.
- (c) Starting conditions for second interim weighing period (Run 2).
Sample weigh boats changed between end of first weighing period and beginning of second weighing period.

APPENDIX C

WEIGH BOAT OXIDATION DATA

APPENDIX C

WEIGH BOAT OXIDATION DATA

In April 1985 oxidation tests were initiated at 135, 150, 165, 180, and 200°C in ovens (low-gamma tests) at approximately 30°C dew point. After 1500 to 1700 h at test temperature, the tests were shut down (end of first "run") and the samples plus weigh boats were weighed. Sample weights were determined by subtracting the pretest tare weights of the respective sample boats. Any weight gain resulting from sample boat oxidation would be included in the weight gain of the fuel sample.

Operational difficulties prevented the tests from restarting until approximately 1 month later. Before the startup of the second run of these tests, 28 new aluminum weigh boats were weighed; the fuel samples were transferred to these boats; and the sample plus weigh boat weights were taken. Thus, the weight of the fuel sample prior to the second run did not include a possible contribution from aluminum oxide formation on the sample boat. Subtracting the sample weights at the end of the first run from their respective sample weights before the beginning of the second run resulted in the following differences in sample weights:

<u>Sample Weight Difference, g</u> <u>(second run minus first run)</u>	<u>No. of Samples</u>
$\leq \pm 0.005$	18
-0.005 to -0.010	6
< -0.01	2
$> +0.006$	2

Eighteen of the 28 samples had insignificant weight differences of $\leq \pm 0.005$ g. For the six samples with a negative difference, there are two possible explanations, i.e., fragment or powder loss during fuel transfer, or weight gain of the sample boats. The former is favored for both spent fuel fragments and nonirradiated pellets that have formed some powder. Differences

in sample weights between the beginning of the second run and end of the first run for nonirradiated pellets that had not fractured into fines or powder after 1500 to 1700 h were:

<u>Temperature, °C</u>	<u>Sample Weight Difference, g (second run minus first run)</u>	<u>No. of Samples</u>
135	0.000	1
150	-0.002	1
165	+0.002	1
180	+0.001	1

These differences are within the accuracy of the balance and suggest that sample boat oxidation is not a significant source of error. In addition, visual examination of sample boats being used in the high-gamma tests after approximately 1700 h at 230°C (20°C dew point) and 2400 h at 190°C (40°C dew point) revealed no visible signs of oxide formation. Based on the evidence, it is concluded that oxidation of the sample boats is insignificant with respect to sample weight.

APPENDIX D

CONTROLLED-ATMOSPHERE SPENT FUEL TESTS
IN HIGH GAMMA FIELD

TABLE D.1. High-Gamma-Field Oxidation Test Matrix
in Hermetically Sealed Ovens

Oven #	Nominal Temperature, °C	Nominal Dew Point, °C
1	150	20
2	150	40
3(a)	170	20
4	190	20
5	190	40
6	135	40
7	150	0
8	190	0
9	210	20
10	135	0
11(a)	170	20
12	230	20

(a) Replicated temperature/dew point combination to provide an estimate of between-oven experimental variation.

TABLE D.2. Pretest Cover Gas Analyses for High-Gamma-Field
Oxidation Tests on Spent Fuel^(a) in Hermetically
Sealed Ovens

Date	Concentration, mol%								NO _x
	CO ₂	Ar	O ₂	N ₂	CO	He	H ₂	CH ₄	
12/18/85	0.02	0.05	21.1	78.4	<0.1	0.36	<0.1	<0.1	NA ^(b)
01/21/85	0.03	0.22	18.5	80.7	<0.1	0.52	<0.01	<0.01	NA
02/19/86	0.02	0.05	20.8	78.4	<0.1	0.74	<0.01	<0.01	NA
03/12/86	0.03	0.03	20.7	79.0	<0.1	0.25	<0.01	<0.01	NA
04/01/86	0.02	0.04	20.9	78.3	<0.1	0.72	<0.01	<0.01	<0.01

(a) Analyses were done when a new source gas was used.

(b) NA = Not Analyzed

TABLE D.3. Cover Gas Analyses for High-Gamma-Field Oxidation Tests on Spent Fuel in Hermetically Sealed Ovens

Oven #	Run # ^(b)	Concentration, mol% ^(a)								
		CO ₂	Ar	O ₂	N ₂	CO	He	H ₂	CH ₄	NO _x
1	1	0.41	0.90	20.2	78.1	<0.1	0.34	<0.01	0.01	0.01
2	1	1.52	0.30	13.6	84.1	<0.1	0.26	0.02	0.12	0.07
2	2	0.14	0.06	20.5	79.0	<0.1	0.33	<0.01	<0.01	NA ^(c)
3	1	1.08	0.09	16.7	81.6	<0.1	0.48	<0.01	0.06	NA
3	2	1.98	0.31	13.4	83.8	<0.1	0.38	<0.01	0.09	0.03
4	1	0.78	0.08	16.7	82.1	<0.1	0.36	<0.01	0.05	NA
4	2	1.11	0.08	16.5	81.9	<0.1	0.31	<0.01	0.06	0.04
4	3	2.01	0.09	12.5	84.9	<0.1	0.34	<0.01	0.12	0.04
5	1	0.80	0.10	16.5	82.2	<0.1	0.28	<0.01	0.04	0.02
5	2	0.95	0.15	15.9	82.3	<0.1	0.58	<0.01	0.05	NA
8	1	1.24	0.08	17.1	81.4	<0.1	0.16	<0.01	0.03	NA
8	2	0.89	0.09	16.1	82.0	<0.1	0.85	<0.01	0.04	0.03
8	3	1.01	0.08	15.7	82.4	<0.1	0.75	<0.01	0.06	0.01
9	2 ^(d)	1.12	0.07	13.9	84.4	<0.1	0.47	<0.01	0.08	NA
9	3	1.18	0.09	14.8	83.3	<0.1	0.49	<0.01	0.06	0.02
11	1	0.83	0.16	17.0	80.5	<0.1	1.45	<0.01	0.04	NA
11	2	1.38	0.12	15.4	82.0	<0.1	1.10	<0.01	0.06	NA
12	1	1.00	0.05	15.1	83.0	<0.1	0.88	<0.01	0.01	0.01
12	2	0.08	0.05	17.6	81.4	<0.1	0.14	<0.01	0.04	NA

(a) Kr and Xe not analyzed for in above runs.

(b) A run is defined as the period of time at test temperature between interim weighings.

(c) NA = Not Analyzed

(d) First run terminated due to a malfunctioning temperature controller.

TABLE D.4. High-Gamma-Field Oxidation Test Raw Data [See footnote (a) for specimen identification]

Sample	Test Temperature, °C	Nominal Dew Point, °C	Burnup, (a) MWd/MTU	Fuel Source	Exposure Time, h	Initial Weight, g	Test Weight, g	Weight Change, g	Weight Change, wt%
10B-1	135	0.0	23.7	SP-F	1702.0	26.492	26.525	0.033	0.125
10B-2	135	0.0	29.5	PB-B	1702.0	22.861	22.871	0.010	0.044
10B-3	135	0.0	27.6	HBRG9T	1702.0	23.496	23.502	0.006	0.026
10B-4	135	0.0	12.5	1A03-3	1702.0	44.660	44.670	0.010	0.022
10B-5	135	0.0	34.1	5D18-3	1702.0	36.802	36.808	0.006	0.016
10B-6	135	0.0	7.9	1A01-2	1702.0	46.400	46.400	0.000	0.000
10B-7	135	0.0	16.7	DTA0103	1702.0	46.000	46.010	0.010	0.022
10B-8	135	0.0	17.0	DTA0103	1702.0	46.520	46.520	0.000	0.000
10B-9	135	0.0	17.2	DTA0103	1702.0	43.270	43.280	0.010	0.023
10B-10	135	0.0	24.2	1C02-4	1702.0	45.930	45.930	0.000	0.000
10B-11	135	0.0	24.2	1C02-4	1702.0	45.870	45.880	0.010	0.022
10B-12	135	0.0	0.0	NI	1702.0	15.746	15.746	0.000	0.000
6B-1	135	40.0	23.7	SP-F	3191.8	27.446	27.577	0.131	0.477
6B-2	135	40.0	29.5	PB-B	3191.8	24.905	24.958	0.053	0.213
6B-3	135	40.0	12.2	1A03-3	3191.8	43.830	43.860	0.030	0.068
6B-4	135	40.0	18.5	1C02-1	3191.8	44.610	44.630	0.020	0.045
6B-5	135	40.0	8.0	1A01-2	3191.8	46.590	46.620	0.030	0.064
6B-6	135	40.0	17.2	DTA0103	3191.8	45.120	45.130	0.010	0.022
6B-7	135	40.0	17.0	DTA0103	3191.8	44.820	44.830	0.010	0.022
6B-8	135	40.0	15.1	0A08-4	3191.8	33.828	33.840	0.012	0.035
6B-9	135	40.0	21.5	0A08-4	3191.8	30.804	30.860	0.056	0.182
6B-10	135	40.0	28.0	0A08-4	3191.8	26.259	26.289	0.030	0.114
6B-11	135	40.0	34.4	0A08-4	3191.8	18.556	18.510	-0.046	-0.248
6B-12	135	40.0	0.0	NI	3191.8	15.603	15.604	0.001	0.006
7B-1	150	0.0	23.7	SP-D	1594.5	21.734	21.918	0.184	0.847
7B-2	150	0.0	29.5	PB-B	1594.5	25.292	25.548	0.256	1.012
7B-3	150	0.0	7.6	1B02-4	1594.5	44.960	45.000	0.040	0.089
7B-4	150	0.0	26.4	HBRG9T	1594.5	24.835	24.682	-0.153	-0.616
7B-5	150	0.0	28.4	HBRG9T	1594.5	22.984	22.424	-0.560	-2.436
7B-6	150	0.0	28.4	HBRG9T	1594.5	25.104	25.164	0.060	0.239
7B-7	150	0.0	26.3	HBRG9T	1594.5	24.293	24.333	0.040	0.165
7B-8	150	0.0	20.8	HBRG9T	1594.5	25.253	25.274	0.021	0.083
7B-9	150	0.0	19.6	HBRG9T	1594.5	24.569	24.617	0.048	0.195
7B-10	150	0.0	15.3	HBRG9T	1594.5	22.978	23.031	0.053	0.231
7B-11	150	0.0	9.9	HBRG9T	1594.5	23.180	23.226	0.046	0.198
7B-12	150	0.0	0.0	NI	1594.5	15.892	15.890	-0.002	-0.013
7B-1	150	0.0	23.7	SP-D	3387.0	21.734	22.047	0.313	1.440
7B-2	150	0.0	29.5	PB-B	3387.0	25.292	25.600	0.308	1.218
7B-3	150	0.0	7.6	1B02-4	3387.0	44.960	45.090	0.130	0.289
7B-4	150	0.0	26.4	HBRG9T	3387.0	24.835	24.720	-0.115	-0.463
7B-5	150	0.0	28.4	HBRG9T	3387.0	22.984	22.466	-0.518	-2.254
7B-6	150	0.0	28.4	HBRG9T	3387.0	25.104	25.208	0.104	0.414
7B-7	150	0.0	26.3	HBRG9T	3387.0	24.293	24.363	0.070	0.288
7B-8	150	0.0	20.8	HBRG9T	3387.0	25.253	25.295	0.042	0.166
7B-9	150	0.0	19.6	HBRG9T	3387.0	24.569	24.658	0.089	0.362
7B-10	150	0.0	15.3	HBRG9T	3387.0	22.978	23.086	0.108	0.470
7B-11	150	0.0	9.9	HBRG9T	3387.0	23.180	23.285	0.105	0.453
7B-12	150	0.0	0.0	NI	3387.0	15.892	15.897	0.005	0.031
1B-1	150	20.0	23.7	SP-D	2129.3	22.028	22.232	0.204	0.926
1B-2	150	20.0	29.5	PB-B	2129.3	24.222	24.296	0.074	0.306
1B-3	150	20.0	29.5	PB-B	2129.3	24.307	24.381	0.074	0.304
1B-4	150	20.0	7.7	1B02-4	2129.3	45.110	45.170	0.060	0.133
1B-5	150	20.0	27.9	HBRG9T	2129.3	23.802	23.838	0.036	0.151
1B-6	150	20.0	28.4	HBRG9T	2129.3	7.260	7.269	0.009	0.124
1B-7	150	20.0	28.4	HBRG9T	2129.3	23.300	23.328	0.028	0.120
1B-8	150	20.0	23.0	HBRG9T	2129.3	24.832	24.849	0.017	0.068
1B-9	150	20.0	18.5	HBRG9T	2129.3	25.175	24.214	-0.961	-3.817
1B-10	150	20.0	17.1	HBRG9T	2129.3	20.092	20.107	0.015	0.075
1B-11	150	20.0	11.7	HBRG9T	2129.3	23.690	23.711	0.021	0.089
1B-12	150	20.0	0.0	NI	2129.3	16.156	16.155	-0.001	-0.006

TABLE D.4. (contd)

Sample	Test Temperature, °C	Nominal Dew Point, °C	Burnup, (a) MWd/MTU	Fuel Source	Exposure Time, h	Initial Weight, g	Test Weight, g	Weight Change, g	Weight Change, wt%
1B-1	150	20.0	23.7	SP-D	2829.3	22.028	22.319	0.291	1.321
1B-2	150	20.0	29.5	PB-B	2829.3	24.222	24.367	0.145	0.599
1B-3	150	20.0	29.5	PB-B	2829.3	24.307	24.377	0.070	0.288
1B-4	150	20.0	7.7	1B02-4	2829.3	45.110	45.210	0.100	0.222
1B-5	150	20.0	27.9	HBRG9T	2829.3	23.802	23.858	0.056	0.235
1B-6	150	20.0	28.4	HBRG9T	2829.3	7.260	7.276	0.016	0.220
1B-7	150	20.0	28.4	HBRG9T	2829.3	23.300	23.346	0.046	0.197
1B-8	150	20.0	23.0	HBRG9T	2829.3	24.832	24.860	0.028	0.113
1B-9	150	20.0	18.5	HBRG9T	2829.3	25.175	24.227	-0.948	-3.766
1B-10	150	20.0	17.1	HBRG9T	2829.3	20.092	20.118	0.026	0.129
1B-11	150	20.0	11.7	HBRG9T	2829.3	23.690	23.726	0.036	0.152
1B-12	150	20.0	0.0	NI	2829.3	16.156	16.160	0.004	0.025
2B-1	150	40.0	23.7	SP-D	1626.2	17.652	17.820	0.168	0.952
2B-2	150	40.0	29.5	PB-B	1626.2	24.759	24.803	0.044	0.178
2B-3	150	40.0	7.8	1B02-4	1626.2	43.910	43.950	0.040	0.091
2B-4	150	40.0	28.4	HBRG9T	1626.2	24.988	25.024	0.036	0.144
2B-5	150	40.0	28.4	HBRG9T	1626.2	24.553	24.585	0.032	0.130
2B-6	150	40.0	24.9	HBRG9T	1626.2	21.216	21.240	0.024	0.113
2B-7	150	40.0	13.4	HBRG9T	1626.2	24.947	24.967	0.020	0.080
2B-8	150	40.0	14.6	OA03-4	1626.2	31.621	31.647	0.026	0.082
2B-9	150	40.0	21.4	OA03-4	1626.2	31.590	31.603	0.013	0.041
2B-10	150	40.0	28.0	OA03-4	1626.2	26.855	26.897	0.042	0.156
2B-11	150	40.0	34.4	OA03-4	1626.2	24.808	24.859	0.051	0.206
2B-12	150	40.0	0.0	NI	1626.2	16.018	16.020	0.002	0.012
3B-1	170	20.0	23.7	SP-D	474.0	20.824	21.154	0.330	1.585
3B-2	170	20.0	29.5	PB-B	474.0	24.797	24.878	0.081	0.327
3B-3	170	20.0	29.5	PB-B	474.0	23.845	23.901	0.056	0.235
3B-4	170	20.0	7.9	1B02-4	474.0	43.990	44.030	0.040	0.091
3B-5	170	20.0	34.0	OA03-4	474.0	24.168	24.218	0.050	0.207
3B-6	170	20.0	23.5	1A01-1	474.0	44.790	44.820	0.030	0.067
3B-7	170	20.0	23.5	1A01-1	474.0	45.140	45.170	0.030	0.066
3B-8	170	20.0	12.5	1A03-3	474.0	46.500	46.530	0.030	0.065
3B-9	170	20.0	19.6	1C02-1	474.0	44.350	43.640	-0.710	-1.601
3B-10	170	20.0	16.0	1C02-1	474.0	44.710	44.730	0.020	0.045
3B-11	170	20.0	8.7	1C02-1	474.0	43.900	43.910	0.010	0.023
3B-12	170	20.0	0.0	NI	474.0	15.873	15.898	0.025	0.158
3B-1	170	20.0	23.7	SP-D	1704.0	20.824	21.391	0.567	2.723
3B-2	170	20.0	29.5	PB-B	1704.0	24.797	25.025	0.228	0.919
3B-3	170	20.0	29.5	PB-B	1704.0	23.845	23.997	0.152	0.637
3B-4	170	20.0	7.9	1B02-4	1704.0	43.990	44.250	0.260	0.591
3B-5	170	20.0	34.0	OA03-4	1704.0	24.168	24.395	0.227	0.939
3B-6	170	20.0	23.5	1A01-1	1704.0	44.790	44.960	0.170	0.380
3B-7	170	20.0	23.5	1A01-1	1704.0	45.140	45.290	0.150	0.332
3B-8	170	20.0	12.5	1A03-3	1704.0	46.500	46.700	0.200	0.430
3B-9	170	20.0	19.6	1C02-1	1704.0	44.350	43.770	-0.580	-1.308
3B-10	170	20.0	16.0	1C02-1	1704.0	44.710	44.840	0.130	0.291
3B-11	170	20.0	8.7	1C02-1	1704.0	43.900	44.010	0.110	0.251
3B-12	170	20.0	0.0	NI	1704.0	15.873	15.908	0.035	0.221
11B-1	170	20.0	23.7	SP-F	491.7	24.332	24.581	0.249	1.023
11B-2	170	20.0	29.5	PB-B	491.7	23.854	23.950	0.096	0.402
11B-3	170	20.0	29.5	PB-B	491.7	23.308	23.384	0.076	0.326
11B-4	170	20.0	27.6	HBRG9T	491.7	22.992	23.038	0.046	0.200
11B-5	170	20.0	12.0	1A03-3	491.7	44.240	44.270	0.030	0.068
11B-6	170	20.0	19.4	1C02-1	491.7	44.560	44.580	0.020	0.045
11B-7	170	20.0	33.7	5D18-3	491.7	35.173	35.204	0.031	0.088
11B-8	170	20.0	8.0	1A01-2	491.7	46.150	46.170	0.020	0.043
11B-9	170	20.0	17.2	DTA0103	491.7	44.490	44.500	0.010	0.022
11B-10	170	20.0	17.2	DTA0103	491.7	43.270	43.280	0.010	0.023
11B-11	170	20.0	24.2	1C02-4	491.7	46.160	46.180	0.020	0.043
11B-12	170	20.0	0.0	NI	491.7	16.333	16.334	0.001	0.006
11B-1	170	20.0	23.7	SP-F	1491.6	24.332	24.764	0.432	1.775

TABLE D.4. (contd)

Sample	Test Temperature, °C	Nominal Dew Point, °C	Burnup, (a) MWd/MTU	Fuel Source	Exposure Time, h	Initial Weight, g	Test Weight, g	Weight Change, g	Weight Change, wt%
11B-2	170	20.0	29.5	PB-B	1491.6	23.854	24.055	0.201	0.843
11B-3	170	20.0	29.5	PB-B	1491.6	23.308	23.477	0.169	0.725
11B-4	170	20.0	27.6	HBRG9T	1491.6	22.992	23.117	0.125	0.544
11B-5	170	20.0	12.0	1A03-3	1491.6	44.240	44.360	0.120	0.271
11B-6	170	20.0	19.4	1C02-1	1491.6	44.560	44.650	0.090	0.202
11B-7	170	20.0	33.7	5D18-3	1491.6	35.173	35.271	0.098	0.279
11B-8	170	20.0	8.0	1A01-2	1491.6	46.150	46.260	0.110	0.238
11B-9	170	20.0	17.2	DTA0103	1491.6	44.490	44.560	0.070	0.157
11B-10	170	20.0	17.2	DTA0103	1491.6	43.270	43.340	0.070	0.162
11B-11	170	20.0	24.2	1C02-4	1491.6	46.160	46.230	0.070	0.152
11B-12	170	20.0	0.0	NI	1491.6	16.333	16.340	0.007	0.043
11B-1	170	20.0	23.7	SP-F	2771.0	24.332	24.986	0.654	2.688
11B-2	170	20.0	29.5	PB-B	2771.0	23.854	24.345	0.491	2.058
11B-3	170	20.0	29.5	PB-B	2771.0	23.308	23.697	0.389	1.669
11B-4	170	20.0	27.6	HBRG9T	2771.0	22.992	23.335	0.343	1.492
11B-5	170	20.0	12.0	1A03-3	2771.0	44.240	44.600	0.360	0.814
11B-6	170	20.0	19.4	1C02-1	2771.0	44.560	44.840	0.280	0.628
11B-7	170	20.0	33.7	5D18-3	2771.0	35.173	35.446	0.273	0.776
11B-8	170	20.0	8.0	1A01-2	2771.0	46.150	46.480	0.330	0.715
11B-9	170	20.0	17.2	DTA0103	2771.0	44.490	44.750	0.260	0.584
11B-10	170	20.0	17.2	DTA0103	2771.0	43.270	43.510	0.240	0.555
11B-11	170	20.0	24.2	1C02-4	2771.0	46.160	46.380	0.220	0.477
11B-12	170	20.0	0.0	NI	2771.0	16.333	16.345	0.012	0.073
8B-1	190	0.0	23.7	SP-E	260.4	26.447	26.673	0.226	0.855
8B-2	190	0.0	29.5	PB-B	260.4	23.638	23.750	0.112	0.474
8B-3	190	0.0	7.9	1B02-4	260.4	40.430	40.450	0.020	0.049
8B-4	190	0.0	33.5	0A03-4	260.4	24.359	24.403	0.044	0.181
8B-5	190	0.0	23.7	1A01-1	260.4	45.300	45.310	0.010	0.022
8B-6	190	0.0	23.5	1A01-1	260.4	44.890	44.900	0.010	0.022
8B-7	190	0.0	12.5	1A03-3	260.4	45.640	45.660	0.020	0.044
8B-8	190	0.0	12.5	1A03-3	260.4	44.030	44.050	0.020	0.045
8B-9	190	0.0	19.8	1C02-1	260.4	44.480	44.490	0.010	0.022
8B-10	190	0.0	18.7	1C02-1	260.4	44.690	44.700	0.010	0.022
8B-11	190	0.0	16.8	1C02-1	260.4	46.770	46.790	0.020	0.043
8B-12	190	0.0	0.0	NI	260.4	16.420	16.424	0.004	0.024
8B-1	190	0.0	23.7	SP-E	638.9	26.447	26.877	0.430	1.626
8B-2	190	0.0	29.5	PB-B	638.9	23.638	23.864	0.226	0.956
8B-3	190	0.0	7.9	1B02-4	638.9	40.430	40.570	0.140	0.346
8B-4	190	0.0	33.5	0A03-4	638.9	24.359	24.552	0.193	0.792
8B-5	190	0.0	23.7	1A01-1	638.9	45.300	45.400	0.100	0.221
8B-6	190	0.0	23.5	1A01-1	638.9	44.890	44.980	0.090	0.200
8B-7	190	0.0	12.5	1A03-3	638.9	45.640	45.760	0.120	0.263
8B-8	190	0.0	12.5	1A03-3	638.9	44.030	44.130	0.100	0.227
8B-9	190	0.0	19.8	1C02-1	638.9	44.480	44.560	0.080	0.180
8B-10	190	0.0	18.7	1C02-1	638.9	44.690	44.770	0.080	0.179
8B-11	190	0.0	16.8	1C02-1	638.9	46.770	46.840	0.070	0.150
8B-12	190	0.0	0.0	NI	638.9	16.420	16.429	0.009	0.055
8B-1	190	0.0	23.7	SP-E	1360.4	26.447	26.979	0.532	2.012
8B-2	190	0.0	29.5	PB-B	1360.4	23.638	23.957	0.319	1.350
8B-3	190	0.0	7.9	1B02-4	1360.4	40.430	40.710	0.280	0.693
8B-4	190	0.0	33.5	0A03-4	1360.4	24.359	24.684	0.325	1.334
8B-5	190	0.0	23.7	1A01-1	1360.4	45.300	45.510	0.210	0.464
8B-6	190	0.0	23.5	1A01-1	1360.4	44.890	45.080	0.190	0.423
8B-7	190	0.0	12.5	1A03-3	1360.4	45.640	45.890	0.250	0.548
8B-8	190	0.0	12.5	1A03-3	1360.4	44.030	44.260	0.230	0.522
8B-9	190	0.0	19.8	1C02-1	1360.4	44.480	44.650	0.170	0.382
8B-10	190	0.0	18.7	1C02-1	1360.4	44.690	44.870	0.180	0.403
8B-11	190	0.0	16.8	1C02-1	1360.4	46.770	46.920	0.150	0.321
8B-12	190	0.0	0.0	NI	1360.4	16.420	16.440	0.020	0.122
8B-1	190	0.0	23.7	SP-E	2347.3	26.447	27.107	0.660	2.496
8B-2	190	0.0	29.5	PB-B	2347.3	23.638	24.221	0.583	2.466

TABLE D.4. (contd)

Sample	Test Temperature, °C	Nominal Dew Point, °C	Burnup, (a) MWd/MTU	Fuel Source	Exposure Time, h	Initial Weight, g	Test Weight, g	Weight Change, g	Weight Change, wt%
8B-3	190	0.0	7.9	1B02-4	2347.3	40.430	41.160	0.730	1.806
8B-4	190	0.0	33.5	0A03-4	2347.3	24.359	24.972	0.613	2.517
8B-5	190	0.0	23.7	1A01-1	2347.3	45.300	45.830	0.530	1.170
8B-6	190	0.0	23.5	1A01-1	2347.3	44.890	45.390	0.500	1.114
8B-7	190	0.0	12.5	1A03-3	2347.3	45.640	46.350	0.710	1.556
8B-8	190	0.0	12.5	1A03-3	2347.3	44.030	44.680	0.650	1.476
8B-9	190	0.0	19.8	1C02-1	2347.3	44.480	44.970	0.490	1.102
8B-10	190	0.0	18.7	1C02-1	2347.3	44.690	45.220	0.530	1.186
8B-11	190	0.0	16.8	1C02-1	2347.3	46.770	47.260	0.490	1.048
8B-12	190	0.0	0.0	NI	2347.3	16.420	16.489	0.069	0.420
4B-1	190	20.0	23.7	SP-E	283.0	27.861	28.150	0.289	1.037
4B-2	190	20.0	29.5	PB-B	283.0	25.136	25.253	0.117	0.465
4B-3	190	20.0	29.5	PB-B	283.0	24.740	24.840	0.100	0.404
4B-4	190	20.0	7.9	1B02-4	283.0	44.680	44.740	0.060	0.134
4B-5	190	20.0	32.4	0A03-4	283.0	24.621	24.671	0.050	0.203
4B-6	190	20.0	23.3	1A01-1	283.0	44.720	44.750	0.030	0.067
4B-7	190	20.0	23.5	1A01-1	283.0	44.760	44.790	0.030	0.067
4B-8	190	20.0	12.5	1A03-3	283.0	43.810	43.850	0.040	0.091
4B-9	190	20.0	19.1	1C02-1	283.0	44.400	44.430	0.030	0.068
4B-10	190	20.0	19.6	1C02-1	283.0	44.430	44.460	0.030	0.068
4B-11	190	20.0	17.5	1C02-1	283.0	48.170	48.190	0.020	0.042
4B-12	190	20.0	0.0	NI	283.0	15.946	15.947	0.001	0.006
4B-1	190	20.0	23.7	SP-E	756.2	27.861	28.318	0.457	1.640
4B-2	190	20.0	29.5	PB-B	756.2	25.136	25.351	0.215	0.855
4B-3	190	20.0	29.5	PB-B	756.2	24.740	24.928	0.188	0.760
4B-4	190	20.0	7.9	1B02-4	756.2	44.680	44.850	0.170	0.380
4B-5	190	20.0	32.4	0A03-4	756.2	24.621	24.555	-0.066	-0.268
4B-6	190	20.0	23.3	1A01-1	756.2	44.720	44.820	0.100	0.224
4B-7	190	20.0	23.5	1A01-1	756.2	44.760	44.860	0.100	0.223
4B-8	190	20.0	12.5	1A03-3	756.2	43.810	43.930	0.120	0.274
4B-9	190	20.0	19.1	1C02-1	756.2	44.400	44.490	0.090	0.203
4B-10	190	20.0	19.6	1C02-1	756.2	44.430	44.520	0.090	0.203
4B-11	190	20.0	17.5	1C02-1	756.2	48.170	48.250	0.080	0.166
4B-12	190	20.0	0.0	NI	756.2	15.946	15.956	0.010	0.063
4B-1	190	20.0	23.7	SP-E	1544.0	27.861	28.440	0.579	2.078
4B-2	190	20.0	29.5	PB-B	1544.0	25.136	25.486	0.350	1.392
4B-3	190	20.0	29.5	PB-B	1544.0	24.740	25.050	0.310	1.253
4B-4	190	20.0	7.9	1B02-4	1544.0	44.680	45.050	0.370	0.828
4B-5	190	20.0	32.4	0A03-4	1544.0	24.621	24.900	0.279	1.133
4B-6	190	20.0	23.3	1A01-1	1544.0	44.720	44.950	0.230	0.514
4B-7	190	20.0	23.5	1A01-1	1544.0	44.760	44.980	0.220	0.492
4B-8	190	20.0	12.5	1A03-3	1544.0	43.810	44.110	0.300	0.685
4B-9	190	20.0	19.1	1C02-1	1544.0	44.400	44.630	0.230	0.518
4B-10	190	20.0	19.6	1C02-1	1544.0	44.430	44.650	0.220	0.495
4B-11	190	20.0	17.5	1C02-1	1544.0	48.170	48.380	0.210	0.436
4B-12	190	20.0	0.0	NI	1544.0	15.946	15.981	0.035	0.219
4B-1	190	20.0	23.7	SP-E	2810.1	27.861	28.593	0.732	2.627
4B-2	190	20.0	29.5	PB-B	2810.1	25.136	25.767	0.631	2.510
4B-3	190	20.0	29.5	PB-B	2810.1	24.740	25.278	0.538	2.175
4B-4	190	20.0	7.9	1B02-4	2810.1	44.680	45.510	0.830	1.858
4B-5	190	20.0	32.4	0A03-4	2810.1	24.621	25.132	0.511	2.075
4B-6	190	20.0	23.3	1A01-1	2810.1	44.720	45.270	0.550	1.230
4B-7	190	20.0	23.5	1A01-1	2810.1	44.760	45.240	0.480	1.072
4B-8	190	20.0	12.5	1A03-3	2810.1	43.810	44.460	0.650	1.484
4B-9	190	20.0	19.1	1C02-1	2810.1	44.400	44.920	0.520	1.171
4B-10	190	20.0	19.6	1C02-1	2810.1	44.430	44.930	0.500	1.125
4B-11	190	20.0	17.5	1C02-1	2810.1	48.170	48.680	0.510	1.059
4B-12	190	20.0	0.0	NI	2810.1	15.946	16.125	0.179	1.123
5B-1	190	40.0	23.7	SP-E	331.0	23.337	23.555	0.218	0.934
5B-2	190	40.0	29.5	PB-B	331.0	24.612	24.714	0.102	0.414
5B-3	190	40.0	22.6	1A01-1	331.0	44.900	44.940	0.040	0.089

TABLE D.4. (contd)

Sample	Test Temperature, °C	Nominal Dew Point, °C	Burnup, (a) MWd/MTU	Fuel Source	Exposure Time, h	Initial Weight, g	Test Weight, g	Weight Change, g	Weight Change, wt%
58-4	190	40.0	23.0	1A01-1	331.0	45.110	45.150	0.040	0.089
58-5	190	40.0	12.5	1A03-3	331.0	45.660	45.750	0.090	0.197
58-6	190	40.0	18.0	1C02-1	331.0	43.790	43.830	0.040	0.091
58-7	190	40.0	10.1	1C02-1	331.0	44.630	44.660	0.030	0.067
58-8	190	40.0	16.0	5D18-3	331.0	36.726	36.767	0.041	0.112
58-9	190	40.0	22.4	5D18-3	331.0	36.875	36.958	0.083	0.225
58-10	190	40.0	27.5	5D18-3	331.0	36.461	36.539	0.078	0.214
58-11	190	40.0	34.4	5D18-3	331.0	37.032	37.140	0.108	0.292
58-12	190	40.0	0.0	NI	331.0	16.109	16.118	0.009	0.056
58-1	190	40.0	23.7	SP-E	735.4	23.337	23.690	0.353	1.513
58-2	190	40.0	29.5	PB-B	735.4	24.612	24.815	0.203	0.825
58-3	190	40.0	22.6	1A01-1	735.4	44.900	45.020	0.120	0.267
58-4	190	40.0	23.0	1A01-1	735.4	45.110	45.210	0.100	0.222
58-5	190	40.0	12.5	1A03-3	735.4	45.660	45.870	0.210	0.460
58-6	190	40.0	18.0	1C02-1	735.4	43.790	43.900	0.110	0.251
58-7	190	40.0	10.1	1C02-1	735.4	44.630	44.700	0.070	0.157
58-8	190	40.0	16.0	5D18-3	735.4	36.726	36.838	0.112	0.305
58-9	190	40.0	22.4	5D18-3	735.4	36.875	37.077	0.202	0.548
58-10	190	40.0	27.5	5D18-3	735.4	36.461	36.649	0.188	0.516
58-11	190	40.0	34.4	5D18-3	735.4	37.032	37.273	0.241	0.651
58-12	190	40.0	0.0	NI	735.4	16.109	16.145	0.036	0.223
58-1	190	40.0	23.7	SP-E	1384.2	23.337	23.773	0.436	1.868
58-2	190	40.0	29.5	PB-B	1384.2	24.612	24.891	0.279	1.134
58-3	190	40.0	22.6	1A01-1	1384.2	44.900	45.100	0.200	0.445
58-4	190	40.0	23.0	1A01-1	1384.2	45.110	45.290	0.180	0.399
58-5	190	40.0	12.5	1A03-3	1384.2	45.660	46.010	0.350	0.767
58-6	190	40.0	18.0	1C02-1	1384.2	43.790	43.980	0.190	0.434
58-7	190	40.0	10.1	1C02-1	1384.2	44.630	44.760	0.130	0.291
58-8	190	40.0	16.0	5D18-3	1384.2	36.726	36.929	0.203	0.553
58-9	190	40.0	22.4	5D18-3	1384.2	36.875	37.213	0.338	0.917
58-10	190	40.0	27.5	5D18-3	1384.2	36.461	36.777	0.316	0.867
58-11	190	40.0	34.4	5D18-3	1384.2	37.032	37.407	0.375	1.013
58-12	190	40.0	0.0	NI	1384.2	16.109	16.179	0.070	0.435
98-1	210	20.0	23.7	SP-E	313.5	23.995	24.303	0.308	1.284
98-2	210	20.0	29.5	PB-B	313.5	23.988	24.182	0.194	0.809
98-3	210	20.0	29.5	PB-B	313.5	23.752	23.962	0.210	0.884
98-4	210	20.0	27.6	HBRG9T	313.5	23.361	23.483	0.122	0.522
98-5	210	20.0	12.5	1A03-3	313.5	44.880	45.060	0.180	0.401
98-6	210	20.0	24.4	HBRG9T	313.5	25.110	25.180	0.070	0.279
98-7	210	20.0	32.6	5D18-3	313.5	36.673	36.798	0.125	0.341
98-8	210	20.0	7.7	1A01-2	313.5	45.670	45.830	0.160	0.350
98-9	210	20.0	17.2	DTA0103	313.5	46.240	46.380	0.140	0.303
98-10	210	20.0	23.8	1C02-4	313.5	45.610	45.730	0.120	0.263
98-11	210	20.0	23.7	1C02-4	313.5	46.280	46.390	0.110	0.238
98-12	210	20.0	0.0	NI	313.5	15.928	15.933	0.005	0.031
98-1	210	20.0	23.7	SP-E	792.3	23.995	24.383	0.388	1.617
98-2	210	20.0	29.5	PB-B	792.3	23.988	24.368	0.380	1.584
98-3	210	20.0	29.5	PB-B	792.3	23.752	24.108	0.356	1.499
98-4	210	20.0	27.6	HBRG9T	792.3	23.361	23.616	0.255	1.092
98-5	210	20.0	12.5	1A03-3	792.3	44.880	45.180	0.300	0.668
98-6	210	20.0	24.4	HBRG9T	792.3	25.110	25.246	0.136	0.542
98-7	210	20.0	32.6	5D18-3	792.3	36.673	36.912	0.239	0.652
98-8	210	20.0	7.7	1A01-2	792.3	45.670	45.960	0.290	0.635
98-9	210	20.0	17.2	DTA0103	792.3	46.240	46.480	0.240	0.519
98-10	210	20.0	23.8	1C02-4	792.3	45.610	45.820	0.210	0.460
98-11	210	20.0	23.7	1C02-4	792.3	46.280	46.470	0.190	0.411
98-12	210	20.0	0.0	NI	792.3	15.928	15.974	0.046	0.289
98-1	210	20.0	23.7	SP-E	1412.1	23.995	24.426	0.431	1.796
98-2	210	20.0	29.5	PB-B	1412.1	23.988	24.491	0.503	2.097
98-3	210	20.0	29.5	PB-B	1412.1	23.752	24.244	0.492	2.071
98-4	210	20.0	27.6	HBRG9T	1412.1	23.361	23.817	0.456	1.952

TABLE D.4. (contd)

Sample	Test Temperature, °C	Nominal Dew Point, °C	Burnup, (a) MWd/MTU	Fuel Source	Exposure Time, h	Initial Weight, g	Test Weight, g	Weight Change, g	Weight Change, wt%
9B-5	210	20.0	12.5	1A03-3	1412.1	44.880	45.530	0.650	1.448
9B-6	210	20.0	24.4	HBRG9T	1412.1	25.110	25.404	0.294	1.171
9B-7	210	20.0	32.6	5018-3	1412.1	36.673	37.229	0.556	1.516
9B-8	210	20.0	7.7	1A01-2	1412.1	45.670	46.280	0.610	1.336
9B-9	210	20.0	17.2	DTA0103	1412.1	46.240	46.800	0.560	1.211
9B-10	210	20.0	23.8	1C02-4	1412.1	45.510	46.080	0.470	1.030
9B-11	210	20.0	23.7	1C02-4	1412.1	46.280	46.700	0.420	0.908
9B-12	210	20.0	0.0	NI	1412.1	15.928	16.084	0.156	0.979
12B-1	230	20.0	23.7	SP-F	61.5	25.681	26.175	0.494	1.924
12B-2	230	20.0	29.5	PB-B	61.5	23.736	24.002	0.266	1.121
12B-3	230	20.0	26.8	HBRG9T	61.5	24.608	24.704	0.096	0.390
12B-4	230	20.0	12.0	1A03-3	61.5	44.300	44.410	0.110	0.248
12B-5	230	20.0	17.7	1C02-1	61.5	36.376	36.431	0.055	0.151
12B-6	230	20.0	8.0	1A01-2	61.5	46.690	46.780	0.090	0.193
12B-7	230	20.0	8.0	1A01-2	61.5	45.880	45.960	0.080	0.174
12B-8	230	20.0	17.2	DTA0103	61.5	45.070	45.120	0.050	0.111
12B-9	230	20.0	24.2	1C02-4	61.5	46.090	46.160	0.070	0.152
12B-10	230	20.0	34.4	0A08-4	61.5	17.394	17.488	0.094	0.540
12B-11	230	20.0	33.9	0A08-4	61.5	15.909	15.991	0.082	0.515
12B-12	230	20.0	0.0	NI	61.5	10.720	10.728	0.008	0.075
12B-1	230	20.0	23.7	SP-F	159.3	25.681	26.226	0.545	2.122
12B-2	230	20.0	29.5	PB-B	159.3	23.736	24.156	0.420	1.769
12B-3	230	20.0	26.8	HBRG9T	159.3	24.608	24.784	0.176	0.715
12B-4	230	20.0	12.0	1A03-3	159.3	44.300	44.510	0.210	0.474
12B-5	230	20.0	17.7	1C02-1	159.3	36.376	36.463	0.087	0.239
12B-6	230	20.0	8.0	1A01-2	159.3	46.690	46.840	0.150	0.321
12B-7	230	20.0	8.0	1A01-2	159.3	45.880	46.020	0.140	0.305
12B-8	230	20.0	17.2	DTA0103	159.3	45.070	45.150	0.080	0.178
12B-9	230	20.0	24.2	1C02-4	159.3	46.090	46.210	0.120	0.260
12B-10	230	20.0	34.4	0A08-4	159.3	17.394	17.538	0.144	0.828
12B-11	230	20.0	33.9	0A08-4	159.3	15.909	16.030	0.121	0.761
12B-12	230	20.0	0.0	NI	159.3	10.720	10.727	0.007	0.065
12B-1	230	20.0	23.7	SP-F	752.8	25.681	26.255	0.574	2.235
12B-2	230	20.0	29.5	PB-B	752.8	23.736	24.280	0.544	2.292
12B-3	230	20.0	26.8	HBRG9T	752.8	24.608	25.135	0.527	2.142
12B-4	230	20.0	12.0	1A03-3	752.8	44.300	45.200	0.900	2.032
12B-5	230	20.0	17.7	1C02-1	752.8	36.376	36.851	0.475	1.306
12B-6	230	20.0	8.0	1A01-2	752.8	46.690	47.410	0.720	1.542
12B-7	230	20.0	8.0	1A01-2	752.8	45.880	46.640	0.760	1.656
12B-8	230	20.0	17.2	DTA0103	752.8	45.070	45.490	0.420	0.932
12B-9	230	20.0	24.2	1C02-4	752.8	46.090	46.800	0.710	1.540
12B-10	230	20.0	34.4	0A08-4	752.8	17.394	17.714	0.320	1.840
12B-11	230	20.0	33.9	0A08-4	752.8	15.909	16.182	0.273	1.716
12B-12	230	20.0	0.0	NI	752.8	10.720	10.785	0.065	0.606
12B-1	230	20.0	23.7	SP-F	1058.1	25.681	26.316	0.635	2.473
12B-2	230	20.0	29.5	PB-B	1058.1	23.736	24.342	0.606	2.553
12B-3	230	20.0	26.8	HBRG9T	1058.1	24.608	25.218	0.610	2.479
12B-4	230	20.0	12.0	1A03-3	1058.1	44.300	45.670	1.370	3.093
12B-5	230	20.0	17.7	1C02-1	1058.1	36.376	37.187	0.811	2.229
12B-6	230	20.0	8.0	1A01-2	1058.1	46.690	48.060	1.370	2.934
12B-7	230	20.0	8.0	1A01-2	1058.1	45.880	47.220	1.340	2.921
12B-8	230	20.0	17.2	DTA0103	1058.1	45.070	46.000	0.930	2.063
12B-9	230	20.0	24.2	1C02-4	1058.1	46.090	47.220	1.130	2.452
12B-10	230	20.0	34.4	0A08-4	1058.1	17.394	17.882	0.488	2.806
12B-11	230	20.0	33.9	0A08-4	1058.1	15.909	16.362	0.453	2.847
12B-12	230	20.0	0.0	NI	1058.1	10.720	10.923	0.203	1.894
12B-1	230	20.0	23.7	SP-F	1329.1	25.681	26.337	0.656	2.554
12B-2	230	20.0	29.5	PB-B	1329.1	23.736	24.362	0.626	2.637
12B-3	230	20.0	26.8	HBRG9T	1329.1	24.608	25.243	0.635	2.580
12B-4	230	20.0	12.0	1A03-3	1329.1	44.300	45.760	1.460	3.296
12B-5	230	20.0	17.7	1C02-1	1329.1	36.376	37.290	0.914	2.513

TABLE D.4. (contd)

Sample	Test Temperature, °C	Nominal Dew Point, °C	Burnup, (a) MWd/MTU	Fuel Source	Exposure Time, h	Initial Weight, g	Test Weight, g	Weight Change, g	Weight Change, wt%
12B-6	230	20.0	8.0	1A01-2	1329.1	46.690	48.200	1.510	3.234
12B-7	230	20.0	8.0	1A01-2	1329.1	45.880	47.330	1.450	3.160
12B-8	230	20.0	17.2	DTA0103	1329.1	45.070	46.170	1.100	2.441
12B-9	230	20.0	24.2	1C02-4	1329.1	46.090	47.330	1.240	2.690
12B-10	230	20.0	34.4	0A08-4	1329.1	17.394	17.906	0.512	2.944
12B-11	230	20.0	33.9	0A08-4	1329.1	15.909	16.384	0.475	2.986
12B-12	230	20.0	0.0	NI	1329.1	10.720	11.000	0.280	2.612
12B-1	230	20.0	23.7	SP-F	1664.2	25.681	26.395	0.714	2.780
12B-2	230	20.0	29.5	PB-B	1664.2	23.736	24.411	0.675	2.844
12B-3	230	20.0	26.8	HBRG9T	1664.2	24.608	25.299	0.691	2.808
12B-4(b)	230	20.0	12.0	1A03-3	1664.2	44.300	45.930	1.630	3.679
12B-5	230	20.0	17.7	1C02-1	1664.2	36.376	37.429	1.053	2.895
12B-6(c)	230	20.0	8.0	1A01-2	1664.2	46.690	48.400	1.710	3.662
12B-7(c)	230	20.0	8.0	1A01-2	1664.2	45.880	47.500	1.620	3.531
12B-8	230	20.0	17.2	DTA0103	1664.2	45.070	46.410	1.340	2.973
12B-9	230	20.0	24.2	1C02-4	1664.2	46.090	47.510	1.420	3.081
12B-10	230	20.0	34.4	0A08-4	1664.2	17.394	17.961	0.567	3.260
12B-11	230	20.0	33.9	0A08-4	1664.2	15.909	16.435	0.526	3.306
12B-12(d)	230	20.0	0.0	NI	1664.2	10.720	11.101	0.381	3.554
12B-1	230	20.0	23.7	SP-F	1996.7	25.681	26.469	0.788	3.068
12B-2	230	20.0	29.5	PB-B	1996.7	23.736	24.392	0.656	2.764
12B-3	230	20.0	26.8	HBRG9T	1996.7	24.608	25.366	0.758	3.080
12B-4(b)	230	20.0	12.0	1A03-3	1996.7	44.300	46.040	1.740	3.928
12B-5	230	20.0	17.7	1C02-1	1996.7	36.376	37.540	1.164	3.200
12B-6(c)	230	20.0	8.0	1A01-2	1996.7	46.690	48.550	1.860	3.984
12B-7(c)	230	20.0	8.0	1A01-2	1996.7	45.880	47.610	1.730	3.771
12B-8	230	20.0	17.2	DTA0103	1996.7	45.070	46.560	1.490	3.306
12B-9	230	20.0	24.2	1C02-4	1996.7	46.090	47.670	1.580	3.428
12B-10	230	20.0	34.4	0A08-4	1996.7	17.394	18.003	0.609	3.501
12B-11	230	20.0	33.9	0A08-4	1996.7	15.909	16.470	0.561	3.526
12B-12(d)	230	20.0	0.0	NI	1996.7	10.720	11.182	0.462	4.310
12B-1	230	20.0	23.7	SP-F	2501.5	25.681	26.612	0.931	3.625
12B-2	230	20.0	29.5	PB-B	2501.5	23.736	24.498	0.762	3.210
12B-3	230	20.0	26.8	HBRG9T	2501.5	24.608	25.478	0.870	3.535
12B-4(b)	230	20.0	12.0	1A03-3	2501.5	44.300	46.180	1.880	4.244
12B-5	230	20.0	17.7	1C02-1	2501.5	36.376	37.700	1.324	3.640
12B-6(c)	230	20.0	8.0	1A01-2	2501.5	46.690	48.720	2.030	4.348
12B-7(c)	230	20.0	8.0	1A01-2	2501.5	45.880	47.770	1.890	4.119
12B-8(e)	230	20.0	17.2	DTA0103	2501.5	45.070	46.740	1.670	3.705
12B-9	230	20.0	24.2	1C02-4	2501.5	46.090	47.890	1.800	3.905
12B-10	230	20.0	34.4	0A08-4	2501.5	17.394	18.073	0.679	3.904
12B-11	230	20.0	33.9	0A08-4	2501.5	15.909	16.527	0.618	3.885
12B-12(d)	230	20.0	0.0	NI	2501.5	10.720	11.255	0.535	4.991
12B-1	230	20.0	23.7	SP-F	3222.8	25.681	26.842	1.161	4.521
12B-2	230	20.0	29.5	PB-B	3222.8	23.736	24.673	0.937	3.948
12B-3	230	20.0	26.8	HBRG9T	3222.8	24.608	25.665	1.057	4.295
12B-4(b)	230	20.0	12.0	1A03-3	3222.8	44.300	46.350	2.050	4.628
12B-5	230	20.0	17.7	1C02-1	3222.8	36.376	37.850	1.474	4.052
12B-6(f)	230	20.0	8.0	1A01-2	3222.8	46.690	48.920	2.230	4.776
12B-7(f)	230	20.0	8.0	1A01-2	3222.8	45.880	47.940	2.060	4.490
12B-8(e)	230	20.0	17.2	DTA0103	3222.8	45.070	46.910	1.840	4.083
12B-9	230	20.0	24.2	1C02-4	3222.8	46.090	48.090	2.000	4.339
12B-10	230	20.0	34.4	0A08-4	3222.8	17.394	18.129	0.735	4.226
12B-11	230	20.0	33.9	0A08-4	3222.8	15.909	16.574	0.665	4.180
12B-12(d)	230	20.0	0.0	NI	3222.8	10.720	11.286	0.566	5.280
12B-1	230	20.0	23.7	SP-F	3961.5	25.681	26.930	1.249	4.864
12B-2	230	20.0	29.5	PB-B	3961.5	23.736	24.745	1.009	4.251
12B-3	230	20.0	26.8	HBRG9T	3961.5	24.608	25.748	1.140	4.633
12B-4(b)	230	20.0	12.0	1A03-3	3961.5	44.300	46.450	2.150	4.853
12B-5	230	20.0	17.7	1C02-1	3961.5	36.376	37.920	1.544	4.245
12B-6(f)	230	20.0	8.0	1A01-2	3961.5	46.690	49.080	2.390	5.119

TABLE D.4. (contd)

Sample	Test Temperature, °C	Nominal Dew Point, °C	Burnup, (a) MWd/MTU	Fuel Source	Exposure Time, h	Initial Weight, g	Test Weight, g	Weight Change, g	Weight Change, wt%
12B-7(f)	230	20.0	8.0	1A01-2	3961.5	45.880	48.060	2.180	4.752
12B-8(f)	230	20.0	17.2	DTA0103	3961.5	45.070	46.980	1.910	4.238
12B-9	230	20.0	24.2	1C02-4	3961.5	46.090	45.760	-0.330	-0.716
12B-10	230	20.0	34.4	0A08-4	3961.5	17.394	18.152	0.758	4.358
12B-11	230	20.0	33.9	0A08-4	3961.5	15.909	16.481	0.572	3.595
12B-12(d)	230	20.0	0.0	NI	3961.5	10.720	11.295	0.575	5.364

- (a) Burnups for Point Beach (PB) and Shippingport (SP) are averages based on power history. Burnups for H. B. Robinson (HBRG9T) determined from burnup analysis of rod and gamma scan plots (ref.: Barner 1985). Burnups for all BWR rods (remainder of rods in this test) determined from gamma scan plots, average rod segment burnup data, and assuming linearity of cesium-137 activity as found for HBR rods (ref.: Barner 1985).
- (b) Approximately 5% (by volume) powder observed.
- (c) Approximately 10-15% (by volume) powder observed.
- (d) Approximately 95-100% (by volume) powder observed.
- (e) <1% (by volume) powder observed.
- (f) Approximately 15-20% (by volume) powder observed.

APPENDIX E

150 TO 250°C NONIRRADIATED UO₂ PELLET TESTS IN AIR

TABLE E.1. Initial Characteristics of Nonirradiated UO_2
Pellets for 150°C Oxidation Tests in Air

Pellet Number	Pellet Age, mo	Density, ^(a) g/cm ³	TD, ^(b) %	Surface Area, cm ²	Weight, g	Surface Condition ^(c)
20	60	10.34	94.34	6.0764	11.8183	0
21	60	10.43	95.16	6.0330	11.7960	0
22	60	10.39	94.80	5.8913	11.3657	0
23	60	10.40	94.89	6.0465	11.8125	0
24	60	10.40	94.89	6.0051	11.6879	0
25	60	10.40	94.89	6.0614	11.8492	0
26	60	10.39	94.80	6.1286	12.0183	0
27	60	10.45	95.35	6.0072	11.7527	0
28	60	10.41	94.98	6.0781	11.9031	0
29	60	10.40	94.89	6.0076	11.6536	0
30	60	10.40	94.89	6.0354	11.7531	0
31	60	10.42	95.07	6.0253	11.7734	0
32	60	10.38	94.71	6.0032	11.6697	0
33	60	10.37	94.62	6.0752	11.8486	0
34	60	10.42	95.07	6.0986	11.9772	0
35	60	10.41	94.98	6.0338	11.7780	0
36	60	10.49	95.71	5.1346	9.3552	0
37	60	10.40	94.89	6.0458	11.8042	0

(a) Geometric density.

(b) Theoretical density.

(c) 0 = as-sintered.

TABLE E.2. Weight Gain (mg) Test Results for 150°C Oxidation Tests in Air

Time at Temperature, h	Pellet Number																	
	20	21	22	23	24	25	26	27	28	29	30	31	32	33	34	35	36	37
121.5	0.3	0.0	0.2	0.1	0.1	0.1	0.0	0.1	-0.2	0.0	-0.2	-0.1	0.0	-0.1	0.1	-0.1	0.0	0.1
313	0.1	0.1	0.2	0.1	0.1	-0.1	-0.2	-0.1	-0.2	-0.1	-0.2	-0.1	0.0	-0.1	-0.1	-0.1	-0.1	0.0
477	0.2	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.0	0.0	-0.1	-0.1	0.0	-0.1	0.0	0.0	0.0	0.0
738	-0.1	-0.2	-0.1	-0.1	-0.1	-0.1	-0.2	-0.1	-0.1	0.1	0.0	0.0	0.1	0.0	0.1	0.1	0.1	0.0
955	0.0	-0.2	-0.2	-0.1	0.0	-0.2	-0.3	-0.2	-0.2	0.0	-0.2	-0.2	0.0	-0.1	0.1	0.0	0.0	-0.2
1,350	-0.4	-0.3	-0.1	-0.1	0.0	0.1	-0.2	0.0	-0.1	0.2	-0.1	0.1	0.1	0.1	0.1	0.0	0.0	0.1
1,509	0.3	0.2	0.2	0.3	0.2	0.2	0.1	0.2	0.1	0.2	0.1	0.0	0.0	0.0	0.1	0.1	0.1	0.1
1,672	-0.2	-0.3	-0.3	0.1	-0.2	0.1	0.0	0.1	-0.1	0.1	0.0	0.1	0.1	0.1	0.2	0.1	0.1	0.1
2,490	-0.2	-0.2	-0.1	-0.1	0.0	-0.1	-0.4	0.0	-0.2	0.1	-0.1	-0.2	0.0	0.0	0.1	0.0	0.1	0.0
2,825	-0.4	-0.6	-0.4	-0.2	-0.3	-0.3	-0.4	-0.2	-0.4	-0.1	-0.2	-0.2	0.0	-0.1	0.0	0.0	-0.1	0.0
3,738	0.0	-0.1	-0.1	0.1	0.0	0.3	0.3	0.2	0.0	0.3	0.2	0.2	0.5	0.2	0.4	0.3	0.4	0.4
4,455	0.0	0.2	0.2	-0.2	-0.1	-0.2	-0.1	-0.1	0.0	-0.2	-0.1	-0.1	-0.4	-0.3	-0.5	-0.3	-0.3	-0.5
5,847	0.6	0.6	0.7	0.5	0.5	0.3	0.3	0.4	0.3	0.4	0.3	0.4	0.3	0.2	0.2	0.2	0.1	0.1
12,039	0.2	0.1	(a)	0.4	0.3	0.2	0.3	(a)	0.2	0.5	0.3	0.3	(a)	0.4	0.6	0.3	(b)	0.5
15,733	0.4	0.4		0.6	0.5	0.5	0.4		0.4	0.7	0.5	0.5		0.5	0.6	0.5		0.5
18,373	0.5	0.5		0.6	0.5	0.5	0.5		0.4	0.6	0.6	0.6		0.5	0.6	0.6		0.6
32,485	1.1	1.1		1.1	1.0	1.0	1.0		1.0	1.2	1.1	1.0		1.0	1.1	0.9		

(a) Removed for leaching tests.

(b) Moved to 200°C oxidation test.

TABLE E.3. Initial Characteristics of Nonirradiated UO_2 Pellets for 150°C Oxidation Tests in Air

Pellet Number	Age, mo	Density, ^(a) g/cm ³	TD, ^(b) %	Surface Area, cm ²	Weight, g	Surface Condition ^(c)
114	0	10.473	95.55	3.6248	5.379	1
117	0	10.466	95.49	3.6049	5.329	1
121	0	10.384	94.74	3.5508	5.172	1
126	0	10.522	96.00	3.5494	5.237	1

(a) Immersion density.

(b) Theoretical density.

(c) 1 = ground pellets, freshly reduced in hydrogen.

TABLE E.4. Weight Gain (mg) Test Results for 150°C Oxidation Tests in Air

Time at Temperature, h	Pellet Number			
	114	117	121	126
695	0	0.2	0.8	0.0
1799	0	0.2	1.0	0.0

TABLE E.5. Initial Characteristics of Nonirradiated UO_2 Pellets for 180°C Oxidation Tests in Air

Pellet Number	Pellet Age, mo	Density, ^(a) g/cm ³	TD, ^(b) %	Surface Area, cm ²	Weight, g	Surface Condition ^(c)
61	9	10.45	96.38	3.5673	5.3005	1
62	9	10.44	96.42	3.6183	5.4056	1
63	9	10.43	96.36	3.6345	5.4368	1
64	9	10.42	96.50	3.6082	5.3773	1
65	9	10.38	95.83	3.5935	5.3207	1

(a) Immersion density.

(b) Theoretical density.

(c) 1 = ground.

TABLE E.6. Weight Gain (mg) Test Results for 180°C Oxidation Tests in Air

Time at Temperature, h	Pellet Number				
	61	62	63	64	65
703	0.3	0.3	0.3	0.2	0.3
1,133	0.9	0.8	0.8	0.8	1.0
1,270	1.0	0.9	0.9	0.9	1.1
1,629	1.0	0.9	0.9	0.9	1.1
2,108	1.5	1.1	1.2	1.1	1.4
2,635	1.9	1.3	1.4	1.5	1.6
3,501	2.7	1.9	2.1	2.2	2.4
4,314	3.4	2.3	2.6	2.7	2.8
4,884	3.7	2.5	2.9	3.0	3.1
5,292	4.3	3.1	3.5	3.6	3.5
5,892	5.0	3.8	4.3	4.5	4.3
6,890	5.7	4.4	4.9	5.2	5.0
7,800	6.7	5.2	5.8	6.5	6.1
8,216	8.5	6.1	6.6	7.2	7.1
9,078	9.2	6.6	7.2	9.0	7.6
9,942	9.9 ^(a)	7.4	7.8	9.8 ^(a)	8.4
10,185		7.9 ^(b)			
10,204	10.5		8.4 ^(a)	10.5	9.0 ^(a)
21,964	17.9		15.1	19.5	20.1

(a) U_3O_8 powder beginning to form.

(b) Pellet removed for microprobe oxygen scans.

TABLE E.7. Initial Characteristics of Nonirradiated UO_2 Pellets for
200°C Oxidation Tests in Air

Pellet Number	Pellet Age, mo	Density, ^(a) g/cm ³	TD, ^(b) %	Surface Area, cm ²	Weight, g	Surface Condition ^(c)
1	2	10.39	94.80	3.7103	5.5773	0
2	2	10.43	95.16	3.6977	5.5662	0
3	2	10.44	95.26	3.6924	5.5652	0
4	2	10.41	94.98	3.7291	5.6254	0
5	2	10.42	95.07	3.6536	5.4642	0
6	2	10.45	95.35	3.7073	5.5962	0
7	2	10.45	95.35	3.7007	5.5833	0
8	2	10.45	95.35	3.6932	5.5689	0

(a) Geometric density.

(b) Theoretical density.

(c) 0 = as-sintered.

TABLE E.8. Weight Gain (mg) Test Results for 200°C Oxidation Tests in Air

Time at Temperature, h	Pellet Number							
	1	2	3	4	5	6	7	8
262.5	0.7	0.8	0.4	1.1	0.5	0.9	0.7	0.7
307.5	1.1	1.1	0.7	1.7	0.8	1.3	1.1	1.1
402	1.2	1.4	0.7	1.9	1.0	1.5	1.2	1.2
454	1.6	1.9	1.1	2.3	1.3	1.8	1.5	1.6
572	1.9	2.4	1.1	2.8	1.6	2.3	1.8	2.0
616	2.4	2.8	1.5	3.3	2.1	2.8	2.2	2.4
664	2.7	3.2	1.6	3.6	2.4	3.1	2.4	2.7
735	2.9	3.5	1.6	3.9	2.4	3.3	2.6	2.8
780	3.2	3.9	1.9	4.4	2.8	3.7	2.9	3.2
923	3.3	4.2	1.9	4.9	3.1	4.1	3.1	3.6
971	3.7	4.6	2.1	5.3	3.4	4.4	3.4	3.8
1067	4.2	5.4	2.5	5.9	4.0	5.0	4.0	4.5
1139	4.7	5.9	3.0	6.6	4.5	5.8	4.5	5.1
1257	5.1	6.5	3.1	7.4	4.8	6.2	4.9	5.6
1326	5.3	6.9	3.1	7.4	4.9	6.3	4.9	5.6
1445	5.9	7.6	3.8	8.5	5.8	7.3	6.0	6.6
1569	6.4	8.2	4.1	9.1	6.3	7.8	6.4	7.2
1661	6.8	8.7	4.4	9.6	6.6	8.3	6.7	7.6
1823	7.1	9.1	4.4	10.0	7.0	8.4	7.0	7.9
1918	7.5	9.8	4.7	10.8	7.4	9.1	7.5	8.6
2084	8.0	10.2	5.0	11.5	7.9	9.6	7.9	8.8
2156	8.2	10.5	5.2	12.0	8.2	10.0	8.2	9.3
2418	8.7	10.6	5.4	12.9	8.6	10.5	8.5	9.7
2584	9.1	11.4	5.7	14.2	9.0	10.9	9.1	10.4
2751	9.4	11.9	5.9	15.0	9.4	11.3	9.4	10.9
2915	10.1	12.8	6.3	16.2	9.7	12.2	9.9	11.6
3221	11.0	14.2	6.6	18.1	10.6	13.7	10.5	12.5
3965	14.3	17.9	8.2	(b)	13.1	(b)	12.6	15.5

(a) U_3O_8 powder beginning to form.

(b) Sample removed from test.

TABLE E.9. Initial Characteristics of Nonirradiated UO_2 Pellets of Mixed Type for 200°C Oxidation Tests in Air

Pellet Number	Pellet Age, mo	Density, ^(a) g/cm ³	TD, ^(b) %	Surface Area, cm ²	Weight, g	Surface Condition ^(c)
36 ^(d)	60	10.48 ^(e)	95.62	5.1346	9.3555	0
26	8	10.44	95.26	3.5929	5.3504	1
36	8	10.40	94.89	3.6471	5.4486	1
37	8	10.47	95.53	3.5599	5.2934	1
38	8	10.50	95.80	3.5972	5.3904	1
39	8	10.42	95.07	3.6221	5.4068	1
40	8	10.47	95.53	3.6236	5.4323	1
E1 ^(f)	22	10.31	94.07	4.1694	6.5577	1
5E1 ^(f)	22	10.54	96.17	4.5164	7.5212	1
10E1 ^(f)	22	10.51	95.89	4.4306	7.1468	1

(a) Immersion density

(b) Theoretical density.

(c) 0 = as-sintered.
1 = ground.

(d) Preoxidized at 150°C in ambient air for 5847 h.

(e) Geometric density.

(f) Pellet E1: 0% Gd_2O_3 .
Pellet 5E1: 5% Gd_2O_3 .
Pellet 10E1: 10% Gd_2O_3 .

TABLE E.10. Weight Gain (mg) Test Results for 200°C Oxidation Tests on Nonirradiated UO₂ Pellets of Mixed Type

Time at Temperature, h	Pellet Number									
	36 ^(a)	26	36	37	38	39	40	E1	5E1	10E1
148	-0.2	0.0	0.2	0.1	0.0	0.1	0.0	0.0	0.0	0.0
218	-0.2	0.0	0.2	0.1	0.0	0.2	0.1	-0.1	-0.1	0.0
622	-0.2	0.3	(b)	(b)	(b)	0.1	0.1	0.0	0.1	0.1
987	0.0	0.4				0.2	0.1	0.1	0.3	0.2
1802	0.8 ^(c)	0.9				0.7	0.5	0.4	1.9	1.2
2546	1.2	1.0				0.8	0.5	0.4	3.4	1.9
2738	1.2	1.0 ^(c)				0.8 ^(c)	0.5 ^(c)	0.4	3.8	2.0
3770	1.7	1.8				1.7	0.7	0.5	6.7	3.6
4370	2.0	2.6				2.7	1.2	0.8	8.6	5.3
4898	2.3	3.4				3.8	1.5	1.1	10.3	7.6
5114	2.4	4.0				4.4	1.5	1.2 ^(c)	11.2	9.1
5543	3.3	5.9				6.7	2.3	1.6	13.7	14.0 ^(c)
6021	3.5	7.4				8.5	2.5	1.6	15.7 ^(c)	20.9 ^(c)
6860	4.0	10.4				12.2	3.2	1.8	18.7	39.3
7026	4.0	11.3				13.2	3.7	1.8	18.7	39.3
7193	4.6	(b)				(b)	(b)	2.4	20.8	48.8
7742	4.8							2.5	24.2	63.5
8726	6.2							4.0	32.6	90.8
9296	6.8							4.3	(b)	(b)
9776	7.6							4.4		

(a) Preoxidized at 150°C in ambient air for 5847 h.

(b) Sample removed from test, continued at 230°C.

(c) U₃O₈ powder beginning to form.

TABLE E.11. Initial Characteristics of Nonirradiated UO₂ Pellets for 200°C Oxidation Tests in Air

Pellet Number	Pellet Age, mo	Density, ^(a) g/cm ³	TD, ^(b) %	Surface Area, cm ²	Weight, g	Surface Condition ^(c)
66	0	10.44	95.26	3.4844	5.1193	1
68	4	10.45	95.35	3.5943	5.3617	1
67	0	10.45	95.35	3.6226	5.4203	1
69	4	10.44	95.26	3.5590	5.2786	1
AS	6	10.34	94.34	3.6920	5.6104	0
70	4	10.41	94.98	3.5670	5.2799	1

(a) Immersion density.

(b) Theoretical density.

(c) 0 = as-sintered.

1 = ground.

Pellets 66 and 67: freshly reduced surfaces.

Pellets 68 and 69: aged surfaces.

Pellet AS: aged centerless ground.

Pellet 70: aged as-sintered.

TABLE E.12. Weight Gain (mg) Test Results for 200°C Oxidation Tests in Air

Time at Temperature, h	Pellet Number				Time at Temperature, h	Pellet Number	
	66	68	67	69		AS	70
187	0.9	0.4	1.3	0.4	92	0.3	0.4
616	4.5	0.4	6.8	0.6	521	1.8	0.5
757	5.5	0.5	8.3	0.9	662	2.5	0.5
924	6.3	0.6	9.9	1.1	829	3.0	0.5
1090	7.5	0.6	11.1 ^(a)	1.2	995	3.7	0.6
1493	10.1	0.7		1.2	1398	4.9	0.8
1683	(a)	0.8		1.5	1588	(a)	0.8
2141		1.7		2.0	2046		1.5
2261		1.5		(a)	2261		1.5
2427		1.4			2333		(a)
2808		1.6					
3800		1.9 ^(a)					

(a) U_3O_8 powder formed and sample removed from test.

TABLE E.13. Initial Characteristics of Nonirradiated UO_2 Pellets for 215°C Oxidation Tests in Air

Pellet Number	Pellet Age, mo	Density, ^(a) g/cm ³	TD, ^(b) %	Surface Area, cm ²	Weight, g	Surface Condition ^(c)
1	4	10.48	95.66	3.605	5.391	0
2	4	10.48	95.63	3.633	5.452	0
3	4	10.53	96.06	3.618	5.4408	0

(a) Geometric density.

(b) Theoretical density.

(c) 0 = as-sintered.

TABLE E.14. Weight Gain (mg) Test Results for 215°C Oxidation Tests in Air

Time at Temperature, h	Pellet Number		
	1	2	3
67	0.3	0.3	0.3
134	1.2	0.8	0.8
206	1.6	1.3	1.3
274	1.7	1.4	1.5
346	2.4	2.2	2.3
413	2.8	2.8	2.7
509	3.3	3.3	3.3
559	3.9	3.9	3.8
606	4.3	4.3	4.4
700	5.1	5.1	5.1
772	5.6	5.6	5.7
842	6.0	6.0	6.1
912	6.5	6.6	6.7
982	7.1	7.1	7.4
1052	7.6	7.6(a)	8.1(a)
1122	8.1	8.0	8.5
1146	8.3	8.1	8.8
1216	8.6(a)	8.5	9.1
1286	8.9	8.8	9.6
1356	9.2	9.1	9.8
1426	9.3	9.3	10.1

(a) U_3O_8 powder beginning to form.

TABLE E.15. Initial Characteristics of Nonirradiated UO_2 Pellets for 230°C Oxidation Tests in Air

Pellet Number	Pellet Age, mo	Density, ^(a) g/cm ³	TD, ^(b) %	Surface Area, cm ²	Weight, g	Surface Condition ^(c)
1	0.5	10.47	95.53	3.6179	5.3978	1
2	0.5	10.42	95.07	3.6791	5.5246	0
3	0.5	10.50	95.80	3.6631	5.5324	0
4	0.5	10.40	94.89	3.6065	5.3413	1
5	0.5	10.48	95.62	3.6038	5.3731	1
6	0.5	10.35	94.43	3.6911	5.5155	0
7	0.5	10.45	95.35	3.5494	5.2530	0
8	0.5	10.49	95.71	3.5987	5.3693	1

- (a) Geometric density.
 (b) Theoretical density.
 (c) 0 = as-sintered.
 1 = ground.

TABLE E.16. Weight Gain (mg) Test Results for 230°C Oxidation Tests in Air

Time at Temperature, h	Pellet Number							
	1	2	3	4	5	6	7	8
24	0.4	0.2	0.2	0.7	0.4	0.3	0.2	0.4
48	0.7	0.3	0.2	1.0	0.5	0.6	0.4	0.8
71	1.6	1.1	1.0	2.0	0.9	1.5	0.8	1.3
95	2.2	1.7	1.7	2.8	1.5	2.7	1.4	2.3
161	4.2	3.7	3.4	4.7	2.5	5.6	2.8	4.3
185	4.6	4.4	4.2	5.5	2.9	6.8	3.4	5.0
210	5.9	5.9	5.2	6.6 ^(a)	3.5	8.2 ^(a)	4.2	5.8
231	6.4 ^(a)	6.7 ^(a)	5.8 ^(a)	7.4	3.9	9.2	4.9 ^(a)	6.5 ^(a)
259	7.1	7.7	6.8	8.6	4.3	10.2	5.8	7.6
301	8.4	9.4	8.0	10.6	4.7	11.8	6.7	8.6
327	9.2	11.0	9.0	12.4	5.5 ^(a)	13.7	7.9	9.8
349	10.1	12.1	10.1	13.6	5.7	14.7	8.4	10.6
370	10.5	13.1	10.8	15.1	6.1	15.5	9.3	11.6
396	11.7	14.1	11.7	16.7	6.7	17.1	10.1	12.4
467	15.3	17.8	14.9	21.6	8.6	21.5	12.6	16.0
490	16.4	19.0	15.9	23.0	9.2	22.7	13.5	17.1
537	18.5	21.5	17.6	25.8	10.6	25.2	15.5	19.5
628	22.8	26.4	21.4	31.7	13.5	29.9	19.2	24.1
677	24.7	28.6	23.1	34.4	14.8	32.2	21.0	26.2
728	26.6	30.8	24.7	37.3	16.3	34.6	22.9	28.3
795	29.3	34.3	27.4	41.1	18.2	38.3	25.3	31.0
870	31.2	36.6	29.1	44.7	19.7	41.0	27.3	33.2
893	32.0	37.4	29.8	46.1	20.2	42.0	28.1	33.9
990	35.4	40.9	32.7	51.8	22.8	46.0	31.3	37.1
1042	37.3	42.7	34.1	54.8	24.2	48.7	32.9	39.0

(a) U_3O_8 powder formation.

TABLE E.17. Initial Characteristics of Nonirradiated UO_2 Pellets for 250°C Oxidation Tests in Air

Pellet Number	Pellet Age, mo	Density, ^(a) g/cm ³	TD, ^(b) %	Surface Area, cm ²	Weight, g	Surface Condition ^(c)
4	1	10.39	94.84	3.3221	4.7282	0
5	1	10.42	95.11	3.4616	5.0476	0
6	1	10.42	95.03	3.708	5.5830	0

(a) Geometric density.

(b) Theoretical density.

(c) 0 = as-sintered.

TABLE E.18. Weight Gain (mg) Test Results for 250°C Oxidation Tests in Air

Time at Temperature, h	Pellet Number			
	4	5	6	7 ^(a)
25	0.3	0.5	0.7	0.5
45	0.9	1	1.4	1.1
63	1.5	1.7	2.3	1.8
81	2.3	2.4	3.3	2.7
98	3.2 ^(b)	3.3 ^(b)	4.6 ^(b)	3.7
122				4.6
140				5.6
157				7.1
193				10.6
213				12.9
230				14.5
275				18.0
299				19.8
397				28.4
487				34.6
577				39.2
667				45.3

(a) Pellet 7 is the average weight gain of pellets 4, 5 and 6.

(b) U_3O_8 powder beginning to form.

APPENDIX F

NONIRRADIATED UO_2 PELLET TESTS IN AIR AND 1% NO_2

TABLE F.1. Initial Characteristics for 215°C Oxidation Test on
Nonirradiated UO₂ Pellets in 1% NO₂

Pellet Number	Pellet Age, mo	Density, (a) g/cm ³	TD, (b) %	Surface Area, cm ²	Weight, g	Surface Condition (c)	NO ₂ in Air, vol%
1(d)	4	10.48	95.66	3.605	5.391	0	0.0
2(d)	4	10.48	95.63	3.633	5.452	0	0.0
3(d)	4	10.53	96.06	3.618	5.4408	0	0.0
4	4	10.54	96.21	3.632	5.4811	0	1.0
5	4	10.42	95.05	3.649	5.4542	0	1.0
6	4	10.52	95.98	3.632	5.4676	0	1.0

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- (a) Geometric density.
 (b) Percent theoretical density.
 (c) 0 = as-sintered.
 (d) Data repeated for comparison.

TABLE F.2. Weight Gain (mg) Test Results for 215°C Oxidation Test
on Nonirradiated UO₂ Pellets in 1% NO₂

Time at Temperature, h	Pellet Number					
	1	2	3	4	5	6
67	0.3	0.3	0.3	0.3	0.6	0.4
134	1.2	0.8	0.8	0.6	1.1	0.5
206	1.6	1.3	1.3	1.0	1.9	0.8
274	1.7	1.4	1.5	1.0	3.1 ^(b)	0.8
346	2.4	2.2	2.3	1.8	6.0	1.5
413	2.8	2.8	2.7	2.6	9.7	2.0
509	3.3	3.3	3.3	5.3 ^(b)	19.6	3.0 ^(b)
559	3.9	3.9	3.8	6.3	20.2	3.8
606	4.3	4.3	4.4	8.6	24.1	5.5
700	5.1	5.1	5.1	13.2	34.4	10.1
772	5.6	5.6	5.7	18.3	42.4	13.3
842	6.0	6.0	6.1	21.8	49.0	17.0
912	6.5	6.6	6.7	26.3	55.4	19.8
982	7.1	7.1	7.4	30.8	63.7	24.4
1052	7.6	7.6 ^(a)	8.1 ^(a)	37.3	73.2	28.9
1122	8.1	8.0	8.5	41.1	80.2	32.7
1146	8.3	8.1	8.8	42.9	82.7	33.7
1216	8.6(a)	8.5	9.1	46.3	88.3	37.2
1286	8.9	8.8	9.6	50.5	94.1	39.7
1356	9.2	9.1	9.8	53.5	99.7	43.0
1426	9.3	9.3	10.1	57.4	100.5	45.4

(a) U₃O₈ powder beginning to form.

(b) Pellet fragmented.

TABLE F.3. Initial Characteristics for 250°C Oxidation Test on
Nonirradiated UO₂ Pellets in 1% NO₂

Pellet Number	Pellet Age, mo	Density, ^(a) g/cm ³	TD, ^(b) %	Surface Area, cm ²	Weight, g	Surface Condition ^(c)	NO ₂ in Air, vol%
1	1	10.30	93.96	3.6607	5.4702	0	1
2	1	10.25	93.56	3.7445	5.6286	0	1
3	1	10.36	94.48	3.7011	5.5935	0	1
4 ^(d)	1	10.39	94.84	3.3221	4.7282	0	0
5 ^(d)	1	10.42	95.11	3.4616	5.0476	0	0
6 ^(d)	1	10.42	95.03	3.708	5.5830	0	0

(a) Geometric density.

(b) Percent theoretical density.

(c) 0 = as sintered.

(d) Data repeated for comparison.

TABLE F.4 Weight Gain (mg) Test Results for 250°C Oxidation Test
in 1% NO₂

Time at Temperature, h	Pellet Number							
	1	2	3	4	5	6	7 ^(a)	8 ^(b)
25	1.2	2.4	0.5	0.3	0.5	0.7	1.4	0.5
45	2.6 ^(c)	4.8 ^(c)	1.1 ^(c)	0.9	1	1.4	2.8	1.1
63				1.5	1.7	2.3	3.6	1.8
81				2.3	2.4	3.3	20.6	2.7
98				3.2 ^(c)	3.3 ^(c)	4.6 ^(c)	30.8	3.7
122							61.2	4.6
140							64.6	5.6
157							77.6	7.1
193							109.1	10.6
213							143.5	12.9
230							150.6	14.5
275							207.2	18.0
299							228.3	19.8
397							272.0	28.4
487							297.2	34.6
577							302.8	39.2
667							308.4	45.3

(a) Pellet 7 is the average weight gain of pellets 1, 2, and 3.

(b) Pellet 8 is the average weight gain of pellets 4, 5, and 6.

(c) U₃O₈ powder beginning to form.

APPENDIX G

NONIRRADIATED UO₂ PELLET CONTROLLED-HUMIDITY
OXIDATION TESTS IN AIR

TABLE G.1. Initial Characteristics of Nonirradiated UO_2 Pellets for 200°C Controlled-Humidity Oxidation Tests in Air

Pellet Number	Pellet Age, mo	Density, (a) g/cm ³	TD, (b) %	Surface Area, cm ²	Weight, g	Surface Condition (c)	Dew Point, °C
112	20	10.574	96.48	3.4840	5.1261	1	-40
113	20	10.586	96.59	3.5641	5.3072	1	-40
115	20	10.568	96.42	3.5172	5.1910	1	-40
116	20	10.547	96.23	3.2904	4.6848	1	-40
118	20	10.568	96.42	3.5204	5.1942	1	-7
119	20	10.581	96.54	3.5726	5.3202	1	-7
120	20	10.593	96.65	3.6016	5.3910	1	-7
122	20	10.601	96.72	3.5607	5.3060	1	-7
123	20	10.538	96.15	3.6372	5.4340	1	20
124	20	10.547	96.23	3.5124	5.1712	1	20
125	20	10.593	96.65	3.6015	5.3858	1	20
128	20	10.579	96.52	3.5753	5.3181	1	20

-
- (a) Immersion density.
 (b) Theoretical density.
 (c) 1 = ground.

TABLE G.2. Weight Gain (mg) Test Results for 200°C Controlled-Humidity Oxidation Tests in Air

Time at Temperature, h	Pellet Number											
	112	113	115	116	118	119	120	122	123	124	125	128
206	0.2	0.1	0.1	0.0	0.3	0.2	0.9	0.2	0.1	0.1	0.0	0.0
760	0.5	0.6	0.5	0.2	0.8	0.9	1.1	0.6	0.8	1.0	0.6	0.5
1234	0.9	1.1	1.1	0.6	1.9	2.2	1.8	1.2	1.6	2.4	1.7	1.2
1707	1.5	2.5	1.6	1.2	3.1	3.6	3.3	2.1	3.1	3.7	3.1	2.2
1800	(a)	(a)	(a)	(a)								
2196	2.5	3.4	2.5	2.4	4.5	5.1	4.6	3.3	5.5	5.9	4.8	3.4
2607	3.7	4.4	3.1	3.4	5.3	6.1	5.8	4.1	6.9(a)	6.7(a)	6.6(a)	4.9(a)
3455	5.3	5.5	4.5	5.6	6.4(a)	7.0(a)	7.5(a)	5.3(a)	9.1	8.2	8.5	7.3
3939	6.7	6.9	6.2	9.9	7.5	8.1	8.8	6.2	11.1	9.5	10.0	9.0

(a) Powder formation.

TABLE G.3. Initial Characteristics of Nonirradiated UO_2 Pellets
for 220°C Controlled-Humidity Oxidation Tests in Air

Pellet Number	Pellet Age, mo	Density, ^(a) g/cm ³	TD, ^(b) %	Surface Area, cm ²	Weight, g	Dew Point, °C
94	0	10.40	94.92	3.597	5.3420	-35
95	0	10.37	94.59	3.596	5.3217	-35
96	0	10.43	95.16	3.455	5.0487	-35
103	18	10.44	95.26	3.495	5.1373	-35
104	18	10.36	94.54	3.602	5.3234	-35
105	18	10.45	95.33	3.658	5.4914	-35
97	0	10.47	95.52	3.629	5.4284	-5
98	0	10.44	95.29	3.589	5.3379	-5
99	0	10.43	95.12	3.527	5.1970	-5
106	18	10.44	95.29	3.473	5.0916	-5
107	18	10.30	93.99	3.598	5.2906	-5
108	18	10.45	95.35	3.611	5.3933	-5
100	0	10.43	95.17	3.514	5.1756	20
101	0	10.44	95.24	3.612	5.3928	20
102	0	10.38	94.75	3.592	5.3172	20
109	18	10.47	95.53	3.571	5.3176	20
110	18	10.46	95.46	3.623	5.4233	20
111	18	10.44	95.24	3.466	5.0755	20

(a) Immersion density.

(b) Theoretical density.

TABLE G.4. Weight Gain (mg/cm²) Test Results for 220°C
Controlled-Humidity Oxidation Tests in Air

Pellet Number	Density, % TD	Dew Point, °C	Time, h						
			196	306	363	473	536	646	756
94	94.92	-35	0.03	0.17	0.28	0.36	0.53	0.80 ^(b)	1.17
95	94.59	-35	0.28	0.50	0.72	1.17 ^(b)	1.97	3.45	5.48
96	95.16	-35	0.12	0.23	0.41	0.41	0.46	0.72	0.84 ^(b)
103	95.26	-35	0.09	0.20	0.34	0.37	0.51	0.74	0.97 ^(b)
104	94.54	-35	0.17	0.34	0.50	0.86 ^(b)	1.19	1.75	2.53
105	95.33	-35	0.16	0.27	0.38	0.44	0.57	0.79	0.96
97	95.52	-5	0.14	0.14	0.22	0.28	0.36	0.52	0.77
98	95.29	-5	0.14	0.14	0.20	0.25	0.33	0.45	0.67
99	95.12	-5	0.17	0.20	0.31	0.40	0.48	0.68	0.88
106	95.29	-5	0.26	0.46	0.66	0.89	1.01	1.30	1.55
107	93.99	-5	0.61	1.11	1.53 ^(b)	2.17	2.86	3.97	5.64
108	95.35	-5	0.19	0.19	0.33	0.44	0.53	0.69	0.94
100	95.17	20	0.23	0.31	0.40	0.65	0.80	1.20	1.54
101	95.24	20	0.25	0.25	0.28	0.36	0.53	0.75	1.02
102	94.75	20	0.25	0.36	0.56	0.84	1.06	1.50	1.84
109	95.53	20	0.34	0.42	0.53	0.76	0.90	1.18	1.46
110	95.46	20	0.22	0.22	0.30	0.47	0.66	1.05	1.30
111	95.24	20	0.20	0.26	0.35	0.52	0.72	1.01	1.36

(a) U₃O₈ powder began to form.

TABLE G.5. Initial Characteristics of Nonirradiated UO_2 Pellets for 200°C and 230°C Controlled-Humidity Oxidation Tests in Air

Pellet Number	Dew Point, °C	Pellet Age, mo	Temperature, °C	Density, ^(a) g/cm ³	TD, ^(b) %	Surface Area, cm ²	Weight, g	Surface Condition ^(c)
19 & 20	Dry ^(d)	6	230	10.58 & 10.57	96.53 & 96.44	6.9329	10.1843	1
21 & 22	23	6	230	10.59 & 10.59	96.62 & 96.62	6.8777	10.0811	1
23 & 24	60	6	230	10.58 & 10.56	96.53 & 96.35	7.2527	10.8455	1
55 & 56	Dry ^(d)	6	200	10.39 & 10.43	94.80 & 95.16	7.2133	10.7338	1
57 & 58	23	6	200	10.42 & 10.31	95.07 & 94.07	7.0780	10.3952	1
59 & 60	60	6	200	10.43 & 10.43	95.16 & 95.16	7.2269	10.7827	1

- ^(a) Immersion density.
^(b) Theoretical density.
^(c) 1 = ground.
^(d) <5 ppm moisture.

TABLE G.6. Weight Gain (mg) Test Results for 200°C and 230°C
Controlled-Humidity Oxidation Tests in Air

Time at Temperature, h	Pellet					
	19 & 20	21 & 22	23 & 24	55 & 56	57 & 58	59 & 60
187.7	1.2	1.2	2.4	0.1	0.3	0.3
283.8	1.5	2.9	6.3	0.7	0.5	0.5
376.6	2.2	5.2	9.21 ^(a)	0.5	0.6	0.9
520.6	3.9 ^(a)	9.3	(b)	0.5	0.7	1.5
705.9	10.7	10.9		0.8	1.2	2.7
872.4	22.4	17.1		0.9	1.4	3.4
1418.9	80.0	34.5 ^(a)		2.0	2.9	7.7

(a) One pellet formed U_3O_8 powder.

(b) Both pellets formed U_3O_8 powder.

APPENDIX H

NONIRRADIATED UO_2 PELLET TESTS IN HIGH GAMMA FIELD

TABLE H.1. Initial Characteristics for Oxidation Test on Nonirradiated Fuel Pellets in Gamma Field of 200,000 R/h in Air at 19°C Dew Point (Static)

Pellet Number	Age, mo	Temperature, °C	Density, (a) g/cm ³	TD, (b) %	Surface Area, cm ²	Weight, g	Surface Condition (c)
130	4	140	10.53	96.11	3.625	5.4475	1
131	4	160	10.54	96.19	3.605	5.2776	1
132	4	180	10.54	96.16	3.551	5.3953	1
27	4	200	10.58	96.57	3.588	5.3576	1
28	4	210	10.58	96.51	3.553	5.2790	1
29	4	220	10.58	96.51	3.588	5.3669	1

- (a) Immersion density.
 (b) Theoretical density.
 (c) 1 = ground.

TABLE H.2. Weight Gain (mg) Test Results for Oxidation Test in Gamma Field and Air at 19°C Dew Point (Static)

Time at Temperature, h	Pellet Number		
	130	131	132
350	0.10	0.10	0.10
700	0.10	0.20	0.20
1250	0.40	0.30	0.30
2000	0.30	0.30	0.60

TABLE H.3. Weight Gain (mg) Test Results for Oxidation Test in Gamma Field and Air at 19°C Dew Point (Static)

Time at Temperature, h	Pellet Number		
	27	28	29
330	0.8	2.7	8.9 ^(a)
660	3.1 ^(a)	10.9 ^(a)	37.5
990	5.3	20.4	77.0
1320	199.9		

- (a) U₃O₈ powder beginning to form.

APPENDIX I

EFFECT OF PELLET SURFACE AREA ON OXIDATION BEHAVIOR OF NONIRRADIATED UO_2 PELLETS AND FRAGMENTS

TABLE I.1. Initial Characteristics of Nonirradiated UO_2 Pellets and Fragments for 230°C Oxidation Tests in Air

Pellet Number	Pellet Age, mo	Density, ^(a) g/cm ³	TD, ^(b) %	Surface Area, cm ²	Weight, g	Surface Condition ^(c)
34	4	10.42	96.07	3.5529 ^(d)	5.2566	1
FR2	4	10.46	96.44	129.5 ^(e)	5.1783	N/A ^(f)
51	4	10.46	96.44	3.6207 ^(d)	5.3867	1
52	4	10.46	96.44	3.5731 ^(d)	5.3203	1
FR5	4	10.46	96.44	130.5 ^(e)	5.2200	N/A

(a) Immersion density.

(b) Theoretical density.

(c) 1 = ground.

(d) Pellet surface area determined from geometric calculation.

(e) Fragment surface area from 8ET method, determined to be 25 cm²/g.

(f) N/A = not applicable.

TABLE I.2. Weight Gain (mg) Test Results for 230°C Oxidation
in Air on Nonirradiated Pellets and Fragments

Time at Temperature, h	Pellet Number				
	34	FR2	51	52	FR5
25.6	0.2	2.1	0.2	0.2	1.8
42.5	0.2	2.4	0.2	0.2	2.2
68.1	0.4	2.8	0.2	0.2	2.4
84.6	0.4	2.8	0.2	0.2	2.4
104.6	0.4	3.0	0.4	0.5	2.9
150.6	0.7	3.5	0.7	0.7	3.3
217.6	1.1	4.1	1.0	1.1	4.1
284.6	1.7	5.3	1.5	1.4	5.2
375.6	2.7 (a)	6.8	2.1	1.7	7.3
441.6	3.7	8.7	2.6	2.0	9.7
507.6	5.0	10.9	3.1	2.2	12.6
577.6	5.9	13.0 (a)	3.5 (a)	2.6 (a)	15.2 (a)
648.6	6.7	14.8	3.8	2.8	17.9
717.6	7.7	17.2	4.1	3.0	20.9
786.6	9.0	19.8	4.6	3.5	24.6
857.6	10.1	23.2	5.0	3.6	27.5
926.6	11.7	27.2	5.6	4.0	33.0
995.6	13.8	33.5	6.6	4.4	39.8
1064.6	17.8	40.6	7.6	5.6	47.1
1133.6	19.7	46.8	8.7	6.5	54.0
1202.6	(b)	53.1	9.7	7.6	61.0
1409.6	-	(b)	11.7	9.9	(b)

(a) U_3O_8 powder beginning to form.

(b) Sample removed from test.

APPENDIX J

OXIDATION TESTS IN AIR OF PNL, CRNL,
AND UKCEGB-BNL NONIRRADIATED UO_2 PELLETS

TABLE J.1. Initial Characteristics of Nonirradiated PNL, CRNL, and UKCEGB-BNL UO₂ Pellet Oxidation Tests in Air

Pellet Number	Source ^(a)	Density, ^(b) g/cm ³	TD, ^(c) %	Surface Area, ^(d) cm ²	Grain Size, ^(e) μm	Weight, g
161	PNL	10.59	96.68	3.571	4.9	5.3277
162	PNL	10.62	96.92	3.570	4.9	5.3440
163	PNL	10.53	96.12	3.621	4.9	5.4099
164	PNL	10.59	96.63	3.617	4.9	5.4374
165	PNL	10.57	96.46	3.631	4.9	5.4462
166	PNL	10.52	95.99	3.591	4.9	5.3290
167	PNL	10.60	96.68	3.536	4.9	5.2453
168	PNL	10.52	95.96	3.564	4.9	5.2774
C1	CRNL	10.59	96.61	12.58	9.8	34.9335
C2	CRNL	10.54	96.20	11.96	9.8	32.5311
C3	CRNL	10.53	96.06	12.43	9.8	34.1980
C4	CRNL	10.52	95.94	11.95	9.8	32.4000
C5	CRNL	10.61	96.80	12.47	9.8	34.6106
C6	CRNL	10.53	96.09	11.95	9.8	32.4542
C7	CRNL	10.60	96.69	12.43	9.8	34.4535
C8	CRNL	10.52	95.98	11.93	9.8	32.3900
B1	UKCEGB-BNL	10.76	98.20	12.72	22.6	21.8834
B2	UKCEGB-BNL	10.80	98.58	12.72	22.6	22.0128
B3	UKCEGB-BNL	10.76	98.18	12.67	22.6	21.7524
B4	UKCEGB-BNL	10.77	98.25	12.71	22.6	21.8429
B5	UKCEGB-BNL	10.77	98.25	11.73	22.6	19.7194
B6	UKCEGB-BNL	10.74	98.01	11.67	22.6	19.5965
B7	UKCEGB-BNL	10.76	98.14	11.83	22.6	19.7543
B8	UKCEGB-BNL	10.75	98.09	11.67	22.6	19.6108

- (a) PNL = Pacific Northwest Laboratory.
 CRNL = Chalk River Nuclear Laboratory.
 UKCEGB-BNL = United Kingdom Central Electricity Generating Board-Berkeley Nuclear Laboratory.
- (b) Immersion density.
- (c) Theoretical density.
- (d) Geometric surface areas determined for each pellet.
 Average pellet dimensions:
 PNL pellets 0.325 in. diameter, 0.382 in. length.
 CRNL pellets 0.56 in. diameter, 0.80 in. length.
 UKCEGB-BNL pellets 0.57 in. diameter (outside), 0.249 in. diameter (inside), 0.60 and 0.54 in. length.
- (e) Average grain intercept.

TABLE J.2. Weight Gain^(a) (mg) of PNL, CRNL, and UKCEGB-BNL Pellets for 200°C Oxidation Tests in Air (0° dew point)

Pellet Number	Time at Test Temperature, h																
	200	400	600	800	1000	1200	1400	1600	1800	2000	2200	2400	2600	2800	3000	3200	3400
163 ^(b)	-0.4	0.2	0.5	0.7	1.1	1.3	1.5	1.8	2.6	3.2	3.3	3.5	4.8	5.7	6.2 ^(c)	6.6	7.2
164 ^(b)	-0.4	0.2	0.5	0.6	1.3	1.3	1.5	1.9	2.9	3.7	3.9	4.4	5.7	6.9	7.4 ^(c)	7.8	8.6
167 ^(d)	0.9	2.2	4.0	5.7	8.0	9.4	9.4	10.8	12.2	13.0	14.0	14.9	16.6 ^(c)	18.5	19.8	23.0	25.5
168 ^(d)	1.1	2.6	4.5	5.9	7.5	8.4	8.5	9.6	10.4	11.2	11.8	12.5	13.9 ^(c)	15.3	15.9	16.9	19.7
C3 ^(b)	-0.1	0.7	1.3	1.7	2.3	2.6	3.0	3.3	4.2	4.9	5.4	6.1	7.4	8.5	9.1	10.2	11.3 ^(c)
C4 ^(b)	-0.1	0.9	1.4	1.6	2.2	2.5	2.8	3.1	3.9	4.7	5.0	5.5	6.6	7.7	8.4	9.8	10.7 ^(c)
C7	0.1	0.8	1.5	1.7	2.5	2.5	2.9	3.2	4.1	4.9	5.1	5.6	6.8	7.9	8.4	9.8	10.5 ^(c)
C8	0.3	1.1	1.9	2.1	3.0	3.4	3.6	4.2	5.2	6.2	6.3	6.9	7.8	8.9	9.7	11.2	12.3 ^(c)
B3 ^(b)	-0.3	0.6	0.7	1.0	1.4	1.4	1.4	1.6	2.4	2.7	2.7	2.9	3.7	4.5	4.8	5.4	6.2
B4 ^(b)	-0.4	0.6	0.7	0.8	1.2	1.2	1.2	1.4	2.1	2.4	2.4	2.4	3.0	3.7	3.8	4.0	4.5
B7	-0.5	0.4	0.6	0.6	0.9	0.9	1.2	1.2	1.7	2.0	2.0	2.0	2.6	3.2	3.2	3.2	3.7
B8	-0.3	0.4	0.7	0.8	1.3	1.3	1.6	1.6	2.1	2.6	2.6	2.6	3.3	4.0	4.1	4.6	5.1

(a) Weight gain (normalized to surface area) of all freshly reduced CRNL, UKCEGB-BNL, and PNL pellets used in Figure 38.

(b) Pellets reduced in hydrogen at 1000°C for 2 h prior to testing.

(c) Power formation detected.

(d) Pellets centerless ground immediately before start of test.

TABLE J.3. Weight Gain^(a) (mg) of PNL, CRNL, and UKCEGB-BNL Pellets for 220°C Oxidation Tests in Air (0° dew point)

Pellet Number	Time at Test Temperature, h														
	200	400	600	800	1000	1200	1400	1600	1800	2000	2200	2400	2600	2800	3000
161 ^(b)	0.2	1.9	5.2	6.9	8.3	11.3 ^(c)	12.7	15.3	16.2	21.7	26.3	29.6	34.7	37.5	40.9
162 ^(b)	-0.2	1.0	1.8	3.7	6.4	7.6 ^(c)	10.0	11.9	15.5	18.5	23.6	26.2	30.3	33.4	36.8
165 ^(d)	0.9	3.2	4.7	9.3 ^(c)	14.3	19.7	25.8	29.4	34.5	37.7	41.9	46.0	48.2	50.7	53.1
166 ^(d)	6.4	12.0	17.4 ^(c)	25.0	31.1	37.9	43.1	49.9	54.8	63.9	70.9	73.3	79.3	82.5	85.2
C1 ^(b)	0.9	3.0	5.7	8.8	13.8	19.8 ^(c)	27.3	36.6	45.6	49.1	65.1	77.6	85.2	90.3	105.0
C2 ^(b)	1.3	3.9	6.9	9.1	10.7	14.0 ^(c)	17.6	20.9	23.4 ^(c)	27.6	31.6	34.1	45.1	50.1	63.0
C5	0.6	1.3	2.2	3.8	5.9	7.7 ^(c)	9.7	12.7	16.1	19.8	25.7	28.5	32.8	36.4	41.7
C6	0.6	2.2	4.6	8.2	11.9	15.3 ^(c)	18.1	23.4	28.7	33.0	39.1 ^(c)	47.0	54.9	59.5	65.4
B1 ^(b)	0.2	0.7	1.4	1.9	3.1	4.3	5.5	6.9	7.6	9.2 ^(c)	10.1 ^(c)	10.7	13.1	14.6	16.0
B2 ^(b)	-0.2	0.2	1.0	2.1	4.2	5.1	6.5	8.8	10.7	13.3 ^(c)	16.0	17.1	18.9	20.3	22.6
B5	0.0	1.4	2.6	4.1	5.9	7.7	8.7	11.1	13.0	16.3 ^(c)	17.6	17.8	19.8	22.1	25.1
B6	0.4	1.4	2.4	3.4	4.8	5.3	7.3	8.6	9.5	10.6 ^(c)	13.1	14.6	16.4	18.4	19.5

(a) Weight gain (normalized to surface area) of all freshly reduced CRNL, UKCEGB-BNL, and PNL pellets used in Figure 39.

(b) Pellets reduced in hydrogen at 1000°C for 2 h prior to testing.

(c) Powder formation detected.

(d) Pellets centerless ground immediately before start of test.

APPENDIX K

TEMPERATURE CHANGE TESTS IN AIR
ON NONIRRADIATED UO₂ PELLETS

TABLE K.1. Initial Characteristics for Nonirradiated UO₂ Pellet
Temperature Change Tests in Air

Pellet Number	Pellet Age, mo	Temperature, °C	Density, ^(a) g/cm ³	TD, ^(b) %	Surface Area, cm ²	Weight, g	Surface Condition ^(c)
B	4	200 (2738 h)	10.58	96.53	3.6236	5.4323	1
B13	4	200 (622 h)	10.52	95.99	3.6471	5.4486	1
B22	4	230 (290 h)					
		200 (987 h)	10.59	96.62	3.5599	5.2934	1
		230 (138 h)					
A13	4	230 (85 h)	10.58	96.53	3.5543	5.2819	1
		200 (2501 h)					
36	60	150 (5847 h)	10.48	95.62	5.1346	9.3555	0
		200 (2546 h)					

(a) Immersion density.

(b) Theoretical density, 100% = 10.96 g/cm³.

(c) 1 = ground.

0 = as sintered.

TABLE K.2. Weight Gain Results for Temperature Change
Tests in Air - Pellet B

Temperature, °C	Total Time, h	Weight Gain		
		mg/cm ²	mg	wt%
200	148	0.03	0.1	0.002
	218	0.03	0.1	0.002
	622	0.03	0.1	0.002
	987	0.03	0.1	0.002
	1802	0.14	0.5	0.009
	2546	0.14	0.5	0.009
	2738(a)	0.14	0.5	0.009

(a) U₃O₈ powder began to form.

TABLE K.3. Weight Gain Results for Temperature Change Tests in Air - Pellet B22

Temperature, °C	Total Time, h	Weight Gain		
		mg/cm ²	mg	wt%
200	148	0.03	0.1	0.002
	218	0.03	0.1	0.002
	622	0.06	0.2	0.004
	987	0.14	0.5	0.009
230	987	0.14	0.5	0.009
	1058	0.26	0.9	0.017
	1125 ^(a)	0.37	1.3	0.024

(a) U₃O₈ powder began to form.

TABLE K.4. Weight Gain Results for Temperature Change Tests in Air - Pellet B13

Temperature, °C	Total Time, h	Weight Gain		
		mg/cm ²	mg	wt%
200	148	0.05	0.2	0.004
	218	0.05	0.2	0.004
	622	0.05	0.2	0.004
230	622	0.05	0.2	0.004
	689	0.11	0.4	0.007
	780	0.19	0.7	0.013
	846	0.37	1.3	0.024
	912 ^(a)	0.59	2.2	0.048

(a) U₃O₈ powder began to form.

TABLE K.5. Weight Gain Results for Temperature Change Tests in Air - Pellet A13

Temperature, °C	Total Time, h	Weight Gain		
		mg/cm ²	mg	wt%
230	26	0.06	0.2	0.004
	42	0.06	0.2	0.004
	68	0.08	0.3	0.006
	85	0.08	0.3	0.006
200	85	0.08	0.3	0.006
	450	0.11	0.4	0.008
	1265	0.23	0.8	0.015
	2009	0.28	1.0	0.019
	2201	0.28	1.0	0.019
	2786(a)	0.32	1.1	0.021

(a) U₃O₈ powder began to form.

TABLE K.6. Weight Gain Results for Temperature Change Tests in Air - Pellet 36

Temperature, °C	Total Time, h	Weight Gain		
		mg/cm ²	mg	wt%
150	5847	0.00	0.0	0.0
200	5847	0.00	0.0	0.0
	5995	0.00	0.0	0.0
	5995	0.00	0.0	0.0
	6065	0.00	0.0	0.0
	6469	0.00	0.0	0.0
	6834	0.00	0.0	0.0
	7649	0.00	0.4	0.004
	8393(a)	0.23	1.2	0.013

(a) U₃O₈ powder began to form.

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