

## EFFECT OF BURNUP LEVEL ON OXIDATION OF LWR SPENT FUEL

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### ABSTRACT

Oxidation tests on bare pressurized water reactor (PWR) and boiling water reactor (BWR) spent fuel fragments under static air cover gas conditions are being conducted in the presence of a  $10^5$  R/h gamma field. Test temperature levels are between 135°C and 230°C. Visual examination of the 230°C BWR specimens revealed that the period of time to form low-density powder increased with increasing burnup level for burnups between 8 and 34 Gwd/MTU. Cumulative weight gain data up to 5 wt%, however, was essentially independent over the same burnup range. The PWR fuel specimens did not form powder after 4000 hours of exposure; thus, powder formation appears to occur earlier on BWR fuels.

### INTRODUCTION

In the United States, dry storage of spent light-water reactor (LWR) fuel at reactor sites is a licensed alternative to storage in pools. Because of the potential for some stored fuel rods to contain cladding breaches, and because the oxidation behavior of spent fuel in air has not been adequately defined, licensed storage atmospheres have thus far been restricted to inert gas. An abnormal event, however, could result in the replacement of the inert storage environment with air; at sufficiently high temperatures, low-density powders are produced during oxidation of spent fuel in air. The volume expansion accompanying the production of the powder could extend existing cladding breaches and could potentially lead to the release of respirable-size powder. Alternatively, possible future fuel storage in air under low-temperature conditions would offer economical benefits and would eliminate the need for monitoring to assure retention of an inert gas atmosphere. For either storage scenario, an understanding of spent fuel oxidation behavior in air is required.

Studies using bare spent fuel fragments at Pacific Northwest Laboratory (PNL) are directed toward supporting the development of air storage criteria by characterizing the oxidation behavior in air of spent LWR fuels under simulated worst-case dry storage conditions. These studies involve the examination of PWR and BWR fuel types, having different burnup levels, after exposure to static air for varying periods at temperatures between 135°C and 230°C. Due to the frequency of the examinations, the tests generally approximate unlimited air conditions; however, extended test periods without replenishment of the air cover gas can lead to limited oxygen conditions. To date, a trend of an increasing period required for powder formation with increasing fuel burnup level has been observed for fuel specimens tested at 230°C. This trend is consistent with observations made by another investigator in which fuel powder formed first at the ends in intentionally defected spent fuel rods, i.e., at the locations where the burnup was lowest.

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### EXPERIMENTAL

The fuels being tested include both BWR and PWR spent fuels and nonirradiated PNL-produced (1)  $UO_2$  pellets. The as-fabricated fuel densities and average grain sizes are listed in Table I. Prior to cutting, all rods, except the Shippingport and Point Beach rods, were gamma scanned using a germanium-drifted gamma-ray detector. Burnup levels of the scanned specimens used in testing were determined by assuming a linear relation between cesium-137 counts and burnup level (2). The burnup levels of the nonscanned specimens were estimated based on power history. The range of burnup levels of the specimens used in this study and examples of specimen matrices are listed in Tables II and III.

Spent fuel oxidation tests are being conducted in 12 stainless steel vacuum ovens that have a maximum operating temperature of 400°C. Each oven contains three shelves that hold a maximum of four fuel samples each. Thus, an oven can hold a maximum of 12 bare fuel samples separately in aluminum sample boats. All oven positions are currently being used; thus, 144 samples comprise the test matrix. The internal oven volume is

TABLE I

Test Spent Fuel Characteristics

Reactor	Grain Size <sup>(a)</sup> (microns)	Density <sup>(b)</sup> (% of TD)
Point Beach	10 <sup>(c)</sup>	NA
Shippingport	7.2 <sup>(d)</sup>	NA
H.B. Robinson	4 to 6 <sup>(d,e)</sup>	92 <sup>(e)</sup>
Quad Cities-1	9.6 to 10.9 <sup>(f)</sup>	96.4 to 95.6
Monticello	11.5 <sup>(f)</sup>	95.4
Nonirradiated	4.9 <sup>(g)</sup>	96.0 to 96.7

TD = Theoretical  $UO_2$  density, 10.96 g/cc.

NA = Not available.

(a) Determined by average grain intercept method on unoxidized spent fuel unless otherwise noted.

(b) As-fabricated fuel pellet density.

(c) From TEM examination (3).

(d) Determined from sibling rod.

(e) From Ref. 2.

(f) Nonirradiated pellet grain size (5).

(g) From Ref. 4.

TABLE II

Oxidation Test Fuel Specimens Used  
in 230°C Test in a Gamma Field of  $1.3 \times 10^5$  R/h

Specimen ID	Reactor	Rod ID	Fuel Type	Burnup <sup>(a)</sup> (Gwd/MTU)
12B-12	NA	NA	NI	0
12B-6	Quad Cities-1	1A01-2	BWR	8.0
12B-7	Quad Cities-1	1A01-2	BWR	8.0
12B-4	Quad Cities-1	1A03-3	BWR	12.0
12B-8	Quad Cities-1	DTA0103	BWR	17.2
12B-5	Quad Cities-1	1C02-1	BWR	17.7
12B-1	Shippingport	SP-F	PWR	23.7 <sup>(b)</sup>
12B-9	Quad Cities-1	1C02-4	BWR	24.2
12B-3	H.B. Robinson	G9T	PWR	26.8
12B-2	Point Beach	PB-B	PWR	29.5 <sup>(b)</sup>
12B-11	Monticello	0A08-4	BWR	33.9
12B-10	Monticello	0A08-4	BWR	34.4

NI = Nonirradiated

- (a) Gamma scan of rod is used to determine burnup level of samples. Assumes a linear relation between cesium-137 counts and burnup (2).  
 (b) Sample assigned the estimated average burnup of the rod because no gamma scan was available.

TABLE III

Oxidation Test Fuel Specimens Used  
in 135°C Test in a Gamma Field of  $1.3 \times 10^5$  R/h

Specimen ID	Reactor	Rod ID	Fuel Type	Burnup <sup>(a)</sup> (Gwd/MTU)
6B-12	NA	NA	NI	0
6B-5	Quad Cities-1	1A01-2	BWR	8.0
6B-3	Quad Cities-1	1A03-3	BWR	12.2
6B-8	Monticello	0A08-4	BWR	15.1
6B-7	Quad Cities-1	DTA0103	BWR	17.0
6B-6	Quad Cities-1	DTA0103	BWR	17.2
6B-4	Quad Cities-1	1C02-1	BWR	18.5
6B-9	Monticello	0A08-4	BWR	21.5
6B-1	Shippingport	SP-F	PWR	23.7 <sup>(b)</sup>
6B-10	Monticello	0A08-4	BWR	28.0
6B-2	Point Beach	PB-B	PWR	29.5 <sup>(b)</sup>
6B-11	Monticello	0A08-4	BWR	34.4

NI = Nonirradiated

- (a) Gamma scan of rod is used to determine burnup level of samples. Assumes a linear relation between cesium-137 counts and burnup (2).  
 (b) Sample assigned the estimated average burnup of the rod because no gamma scan was available.

$0.042 \text{ m}^3$ . Temperature is monitored using a chromel-alumel thermocouple positioned between the sample boats on each shelf. Nominal test temperatures are 135°, 150°, 170°, 190°, 210°, and 230°C.

Gamma radiation in a spent fuel storage cask is expected to generate gaseous products in the test atmosphere as a result of radiolysis. These chemically active gases may in turn increase the oxidation rate of spent fuel. To simulate the gamma radiation field under storage conditions, a CsCl gamma-radiation source was placed in front of each oven door while the samples were at test temperature. Dose rates of the sources are approximately  $1.3 \times 10^5$  ( $\pm 14\%$ ) R/h at 0.30 m, which is the approximate distance of the fuel samples from the source.

To prepare fuel for testing, a 3.8- to 4.4-cm-long section was cut from a designated fuel rod location using either a diamond wafering or aluminum oxide saw blade. Archive specimens and specimens for further characterization (e.g., ceramography, electron microscopy) were removed from selected locations from the rod during cutting and stored in ambient air. The fuel to be used for testing was then removed from the cladding. Because the startup of an oven required specimens from several different fuel rods, clad fuel sections were placed in labeled vials and stored within five days after cutting in nitrogen until all samples 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 and the plus No. 16 mesh particles were placed in aluminum sample boats that had been tarred to  $\pm 0.001$  g. The sieving process removed fuel fines generated by cladding removal operations that might otherwise be confused with fines and/or powder produced during fuel oxidation testing. For each test temperature, nonirradiated PNL-produced  $\text{UO}_2$  pellets (1,4) were included in each oven. All sample boats with fuel were weighed to within  $\pm 0.001$  g.

Eleven spent fuel specimens were removed from nitrogen storage for each oven, decladded, and prepared for testing as described above. The typical size of the fragments prior to testing was approximately 5 to 10 mm on edge (1/4 to 1/3 section of a pellet) for fuel with relatively low burnup ( $< 20$  Gwd/MTU), 4 to 6 mm on edge for fuel with intermediate burnup (approximately 20 to 25 Gwd/MTU), and 4 mm or less for the highest burnup fuel ( $> 25$  Gwd/MTU). The lower burnup fuel was easier to remove from the cladding; thus, the larger fragment sizes may be partly due to less fracturing as a result of decladding. Well-characterized nonirradiated PNL-produced  $\text{UO}_2$  pellets (typically three) were included in the twelfth sample boat in each oven. All sample boats with fuel were weighed twice to within  $\pm 0.001$  g ( $\pm 0.01$  g if the total weight was 60 g or over). Differences in weight of a single specimen 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 clad specimen. Starting weights of the nonirradiated pellets were approximately 16 g (each pellet weighed approximately 5.3 g).

Sample boats containing the fuel specimens were carefully placed on preselected shelves. The oven door was sealed and the oven was evacuated to a pressure of approximately 3.4 kPa absolute pressure using a mechanical pump. After evacuation, the test oven was filled with air until a pressure of approximately 109 kPa absolute pressure was obtained. Evacuation and air back-filling were repeated to purge the oven chamber of possible gaseous contaminants and excess water. The cover gas (commercial-grade extra-dry air) simultaneously flowed through a humidification tank (where moisture evaporated into the gas stream) and a humidification tank bypass. By adjustment of the flow rates of the wet and dry gas streams, controlled moisture levels (0°, 20°, or 40°C nominal dew point) were obtained. Once the correct dew point was obtained, the cover gas was allowed to flow for a minimum of three oven volume exchanges. A static test atmosphere within the oven was obtained by closing an inlet header valve and outlet header cap screw.

While an oven was heated to the desired testing temperature, periodic venting through the outlet header cap screw was performed to prevent overpressurization and gasket blowout. The final oven pressure at test temperature was approximately 109 to 112 kPa absolute

pressure. During the tests, the temperature level of each oven shelf and the oven pressure were recorded daily.

Prior to oven shutdown, a 500-mL sample of the static test cover gas was collected for mass spectrometric analysis. After each oven cooled, the samples were removed and weighed twice to within  $\pm 0.001$  g. Using a hot cell periscope, each specimen was carefully inspected for color and luster, size distribution, the presence or absence of cracks, and in particular the presence or absence of powder. The formation of powder could be detected by adherence of the powder to the bottom of the sample boat after the boat had been tilted at approximately a  $45^\circ$  angle. Powder volume was estimated by visual examination. Specimens were returned to the oven and the oven was restarted with a fresh cover gas as previously described.

Only test results for the  $230^\circ\text{C}$  temperature level are presented below because the specimens at this temperature are in the most advanced stages of powder formation. Spent fuel specimens at the second highest temperature level ( $210^\circ\text{C}$ ) are only beginning to show signs of powder formation. A total of three PWR specimens, eight BWR specimens, and one sample of nonirradiated PNL-produced pellets comprised the  $230^\circ\text{C}$  test (Table II). Test specimens from the lowest test temperature of  $135^\circ\text{C}$  are listed for comparison in (Table III).

#### RESULTS AND DISCUSSION

Weight gain behaviors at  $230^\circ\text{C}$  for all 12 specimens are plotted as a function of time in Figs. 1-3. Below exposures of 750 hours, the weight gain behaviors of the three PWR fuels were different from each other. The initial rates of weight gain varied by about a factor of five (Fig. 1). This behavior may be due to undefined differences in microstructure and/or possibly chemical composition, but did not correlate with grain size (Table I). Between 750 and 2000 hours of exposure, the rates of weight gain were significantly reduced with respect to the initial rates and were similar for the three specimens; the level of weight gain increased to approximately 3 wt% over this time period. For the exposure period between approximately 2000 and 4000 hours, the rates of weight gain initially increased and then decreased approximately parabolic-

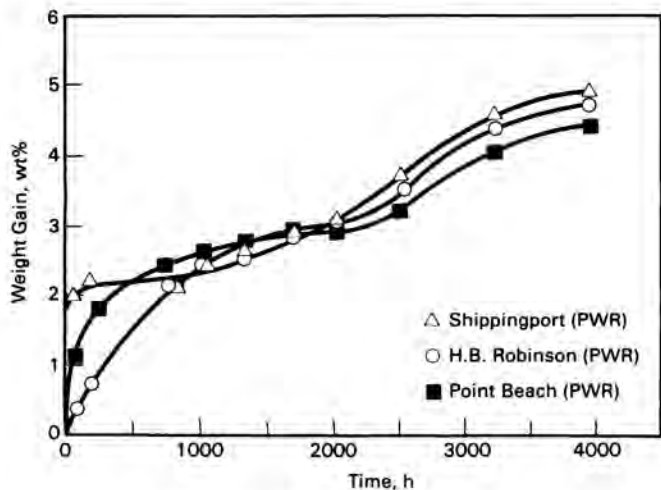


Fig. 1. Weight Gain Behavior of PWR Fuels Tested at  $230^\circ\text{C}$ ,  $20^\circ\text{C}$  Dew Point.

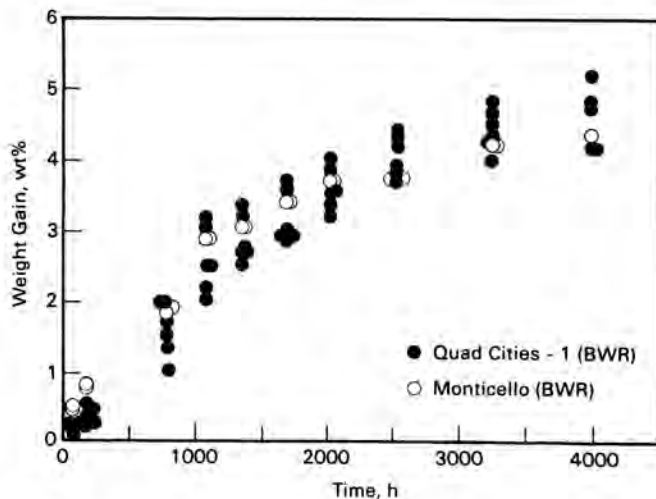


Fig. 2. Weight Gain Behavior of BWR Fuels Tested at  $230^\circ\text{C}$ ,  $20^\circ\text{C}$  Dew Point.

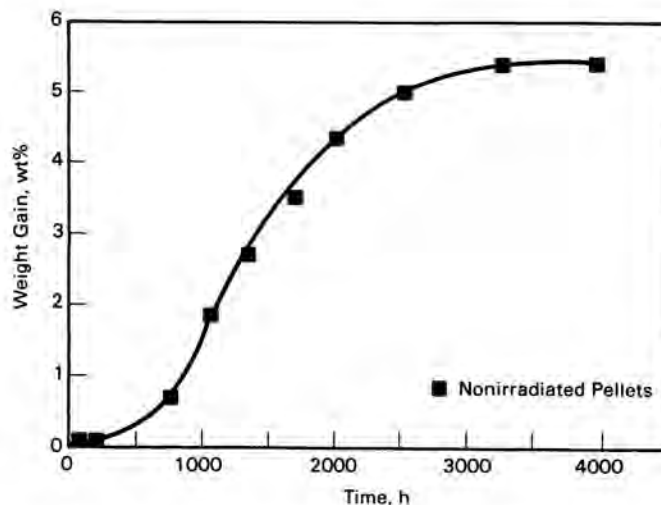


Fig. 3. Weight Gain Behavior of PNL-Produced (1,4) Nonirradiated Pellets at  $230^\circ\text{C}$ ,  $20^\circ\text{C}$  Dew Point.

ally, suggesting behavior indicative of a bulk diffusion-controlled reaction (6). Note that in all PWR fuels tested, the level of weight gain after approximately 4000 hours of exposure exceeded 4 wt%, which, on the average, corresponds to greater than full conversion of  $\text{UO}_2$  to  $\text{U}_3\text{O}_8$ . Apparently, the fuel is proceeding to a higher oxidation state, presumably to  $\text{UO}_3$ .

The total weight gain is attributed to oxidation of the specimen because no significant weight change resulted for an empty aluminum sample boat after approximately 2200 hours of exposure at  $230^\circ\text{C}$  test conditions. Also, no visible degradation of the actual sample boats was observed.

Mass spectrometer analyses of the cover gas prior to shutdown of the  $230^\circ\text{C}$  test oven revealed that the oxygen level varied between about 5 and 19 mole%. In most cases, the oxygen level did not drop below 15 mole%; however, for exposure time periods ending at approximately 750 and 1100 hours, the oxygen concentrations were 4.9% and 8.6%, respectively. The low oxygen

concentrations were attributed to a delay in oven shut-down and to the relatively high rates of oxidation of both the spent and nonirradiated fuels occurring over these time periods.

In general, each BWR fuel specimen exhibited similar weight gain behavior with exposure period (Fig. 2). Below 1100 hours, the cumulative weight gain may have been reduced somewhat due to the previously mentioned low oxygen concentrations, particularly at approximately 750 hours of exposure. For exposure times above 1100 hours, the weight gain behavior was approximately parabolic in shape, suggesting a bulk diffusion-controlled oxidation process. Similar to the PWR fuels, the weight gain of most of the BWR fuels exceeded 4 wt% after approximately 4000 hours of exposure.

The weight gain behavior for nonirradiated pellets was sigmoidal with exposure time (Fig. 3). The sigmoidal shape of weight gain with exposure time for nonirradiated  $UO_2$  has been observed by others at 250°C (7).

For the BWR fuels, weight gains of up to 5% were essentially independent of the burnup level of the specimens from 8 to 34 GWd/MTU at a temperature of 230°C (Fig. 4). This is consistent with results from analysis (8) of oxidation data corresponding to different burnup levels (9). Although the PWR fuels in the 230°C test appeared to follow the same trend, the range of burnup (24 to 30 GWd/MTU) was too narrow to draw similar conclusions.

Because of operational difficulties, visual observations of 230°C test specimens were not recorded prior to 1664 hours of exposure; pretest specimens were not photographed. The condition of specimens at the lowest test temperature of 135°C with an exposure time of 3192 hours approximates the condition of the pretest specimens at 230°C. Both PWR and BWR specimens covering a burnup range similar to the 230°C test specimens were present in the 135°C test group (Table III). The 135°C group of specimens showed no sign of powder forming on any of the fuel specimens, including the nonirradiated  $UO_2$  pellets (Fig. 5). Weight gain levels of these specimens were 0.2 wt% or below except for the Shippingport specimen, which was below 0.5 wt%.

In general, the visual examinations of the 230°C group 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

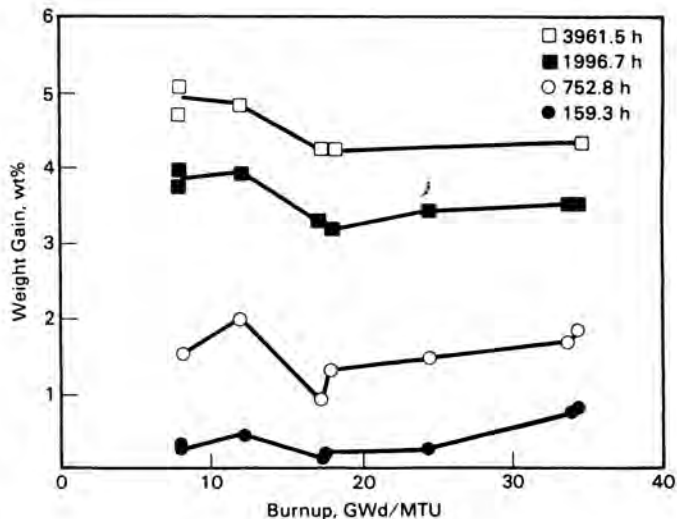
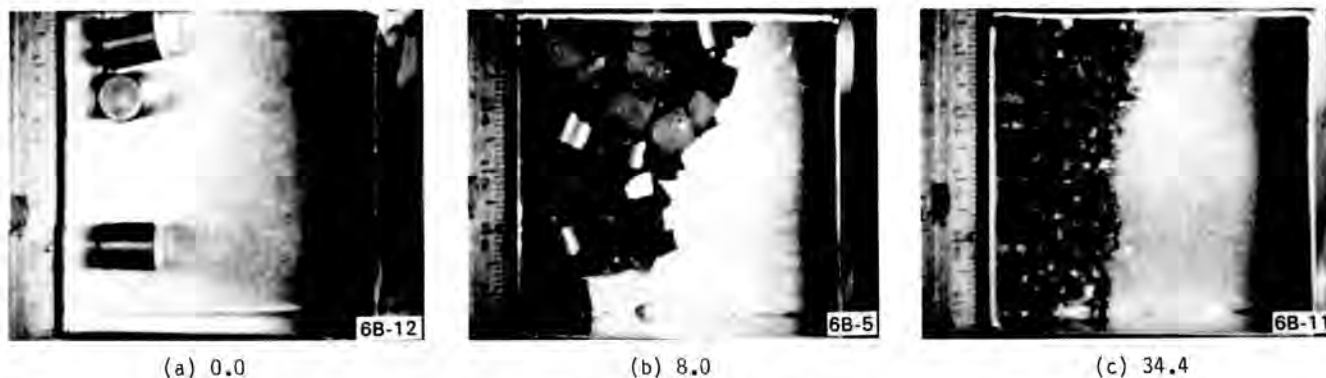


Fig. 4. Weight Gain as a Function of Burnup Level for BWR Fuels Tested at 230°C. Note: Data points at 24.2 and 33.9 GWd/MTU burnup after 3961.5 hours of exposure are omitted because fuel spillage was suspected.

the fragment surfaces to a dull sheen. Also, deep open cracks and/or multiple hairline cracks in the fragments were usually observed when powder had formed. The disappearance of the sharp edges was observed when large quantities of powder were present (20% to 30%); such a fragment, removed for future transmission electron microscopic analysis, easily broke into several pieces during handling.

After the initiation of powder formation and after a particular exposure time, the 230°C group revealed a trend of increasing volume of powder with decreasing burnup level of the specimens. The estimated amounts of powder formed as a function of time are given in Table IV. After 1664 hours of exposure, an estimated 95% (by volume) of the nonirradiated pellets were converted to powder, 10% to 15% powder was observed on fragments at 8.0 GWd/MTU burnup level, and less than 5% powder was observed at the 12.0 GWd/MTU burnup level. The period required for powder formation on the specimens with burnup levels of 12.0 GWd/MTU or less was therefore less than 1664 hours. Powder started to form on the fuel fragments at 17 GWd/MTU burnup after 1664 hours of exposure, and no powder was observed on



(a) 0.0 (b) 8.0 (c) 34.4

Fig. 5. Example of LWR Spent Fuel Exposed 3192 Hours at 135°C in Air (40°C dew point). (Values below photos are burnup values in GWd/MTU.)

TABLE IV

Estimated Volume Percent Powder Formed on Bare Fuel Specimens as a Function of Exposure Time at 230°C

Sample ID	Burnup (Gwd/MTU)	Exposure Time (h)					
		1664 <sup>(a)</sup>	1997	2502	3223	3962	4500 <sup>(b)</sup>
12B-12 (NI)	0.0	95	95	100	100	100	100
12B-6 (BWR)	8.0	10 to 15	10 to 15	10 to 15	15 to 20	15 to 20	20 to 30
12B-7 (BWR)	8.0	10 to 15	10 to 15	10 to 15	15 to 20	15 to 20	20 to 30
12B-4 (BWR)	12.0	<5	5	5	5	5	20 to 30
12B-8 (BWR)	17.2	0	0	<1	<1	<1	5
12B-5 (BWR)	17.7	0	0	0	0	0	0
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

(a) Visual observations not conducted below 1664 hours.

(b) Estimated exposure time.

specimens at higher burnups. Examination of specimens after 1997 hours of exposure (Fig. 6) showed no significant changes in the state of the fuel specimens observed at 1664 hours.

The powder volume for specimens that previously had powder present after 1664 hours of exposure increased with an increased exposure period. After approximately 4500 hours of exposure at 230°C (Fig. 7), powder was observed on all the BWR fuel specimens with the exception of one at 17.7 Gwd/MTU. These results show that for the BWR fuel tested, the period required for powder formation on bare fuel fragments with burnup levels between 17 and 34 Gwd/MTU increases with increasing burnup. This trend also appears to be valid below burnups of 17 Gwd/MTU assuming that the greatest volume of powder present in a specimen indicates the lowest time for powder formation.

Increased time for powder formation as a function of burnup was observed by Gilbert et al. (10) for a variety of LWR fuels under different test conditions at temperatures between 229°C and 283°C. Einziger (11) observed a similar trend for Turkey Point nonirradiated pellets and Turkey Point PWR bare spent fuel fragments at 295°C.

Powder has not been observed on any of the PWR fuel specimens after 4500 hours of exposure; the period required for powder formation on the PWR fuels appears to be greater than for the BWR fuels. This is not solely due to burnup differences because the BWR burnup range of 8.0 to 34.4 Gwd/MTU brackets the narrower PWR burnup range.

Tests on an intentionally defected BWR full-length spent fuel rod were conducted in unlimited air by Olsen (12). The average assembly burnup level was 12 to 13 Gwd/MTU. The rod was subjected to a gradually decreasing temperatures starting at 230°C. In these tests, cladding crack propagation and fuel rod diametral enlargement due to low-density powder formation occurred at the rod-end-defect location (lowest burnup in the rod) after 7200 hours of exposure when the temperature had decreased to 217°C. There was no evidence of cladding enlargement at the rod-center-defect site for the same exposure period. The results of this whole-rod test are qualitatively consistent with the results from this investigation on fuel pellet fragments at 230°C, in which low-density powder formed at an earlier time with decreasing burnup for BWR-type fuel.

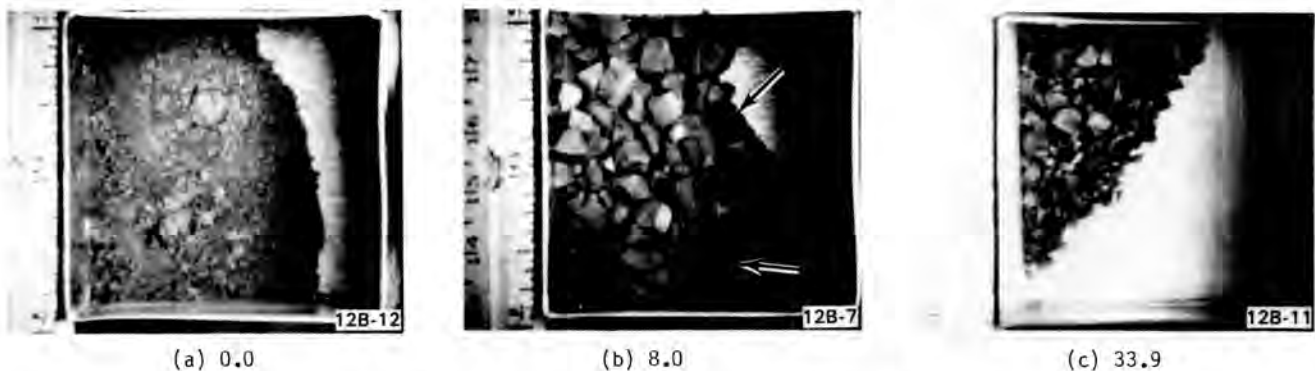


Fig. 6. Example of LWR Spent Fuel Exposed 1997 Hours at 230°C in Air (20°C dew point). (Values below photos are burnup values in Gwd/MTU; arrow in photos show location of powder.)

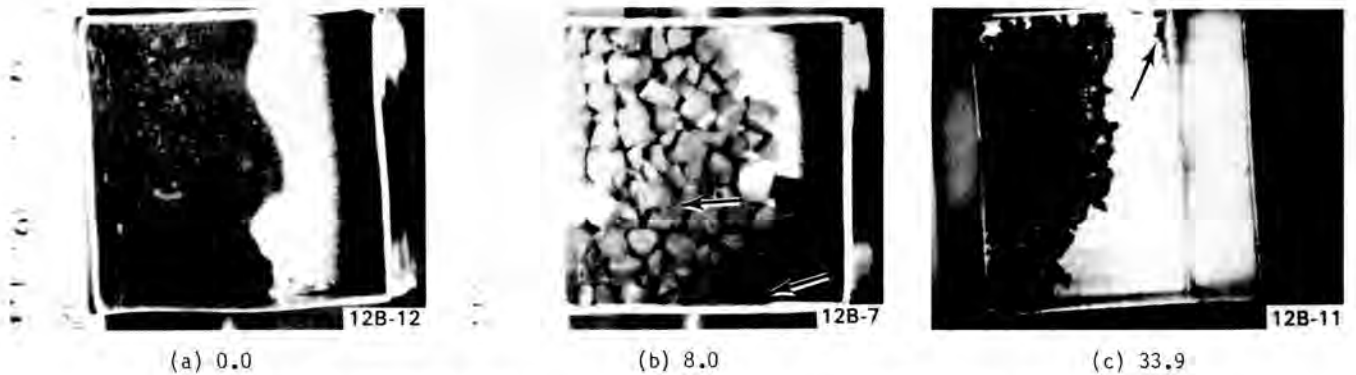


Fig. 7. Example of LWR Spent Fuel Exposed 4500 Hours at 230°C in Air (20°C dew point). (Values below photos are burnup values in Gwd/MTU; arrows in photos show location of powder.)

### CONCLUSIONS

1) The weight gains of PWR fuel from three source reactors were essentially identical after 4000 hours of exposure in air at 230°C. Overall weight gains after 4000 hours of exposure were in excess of 4 wt%.

2) For a given exposure period, the cumulative weight gain of the BWR fuels was independent of the burnup level between 8 and 34 Gwd/MTU. The burnup range was too narrow to determine if this was the case for PWR fuels.

3) Visual examinations conducted on bare BWR spent fuel fragments from the Quad Cities-1 and Monticello reactors show that the period required to form powder at 230°C is an inverse function of burnup for the burnup range between 8 to 34 Gwd/MTU, i.e., the time period for powder formation increased with increasing burnup.

4) For the BWR and PWR fuels tested, the period required to form powder at 230°C is shorter for the BWR fuels. This is not due to a burnup difference between the BWR and PWR fuels because the BWR burnup range of 8.0 to 34.4 Gwd/MTU brackets the narrower PWR burnup range.

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