AN EXPERIMENTAL PROGRAM TO DETERMINE MAXIMUM TEMPERATURES FOR DRY STORAGE OF SPENT FUEL

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ABSTRACT

Although air is used as a cover gas in some dry storage facilities, other facilities use inert cover gases that must be monitored to assure inertness of the atmosphere. Thus, qualifying air as a cover gas is attractive for the dry storage of spent fuels.

At sufficiently high temperatures, air can react with spent fuel (UO_2) at the site of cladding breaches that formed during reactor irradiation or during dry storage. The reaction rate is temperature dependent; hence, the rates can be maintained at acceptable levels if temperatures are low.

Tests with spent fuel are being conducted at Pacific Northwest Laboratory (PNL) to determine the allowable temperatures for storage of spent fuel in air. Tests performed with nonirradiated UO_2 pellets indicated that moisture, surface condition, gamma radiation, gadolinia content of the fuel pellet, and temperature are important variables. Tests were then initiated on spent fuel to develop design data under simulated dry storage conditions.

Tests have been conducted at 200°C and 230°C on spent fuel in air and 275°C in moist nitrogen. The results for nonirradiated UO_2 and published data for irradiated fuel indicate that above 230°C oxidation rates are unacceptably high for extended storage in air. Air tests at temperatures from 135 to 230°C and moist nitrogen tests from 300 to 400°C with spent fuel will be conducted between 135°C and 230°C for approximately three years to enable reliable extrapolations to be made for extended storage in air and inert gases with oxidizing constituents.

INTRODUCTION

Nuclear utility spent fuel storage pools were never designed to provide permanent storage of spent nuclear fuel assemblies. Pool storage capacity shortages will soon occur for some utilities.[1] By law (the Nuclear Waste Policy Act of 1982), the utilities are required to provide interim storage until 1998, at which time the federal government will provide a storage facility and/or a licensed federal repository. Dry spent fuel storage demonstrations are being initiated in the United States to support licensing.[2,3,4] Dry storage is licensed in the Federal Republic of Germany, Canada, and Switzerland.

In an oxidizing atmosphere, UO_2 fuel reacts to form U_3O_8 powder with a temperature-time dependency, i.e., the higher the temperature the faster the reaction will occur. The density reductions that occur during this reaction can create internal pressures and cladding strain, which can propagate existing cracks or form new cracks in the fuel cladding. The split cladding could permit release of the U_3O_8 resulting in contamination of the storage system. To prevent this fuel degradation by oxidation, monitored retrievable storage (MRS) and interim storage concepts are designed to store fuel in a nonoxidizing atmosphere.[5]

Regulatory and design criteria (Code of Federal Regulations 1984)[6] for dry storage of spent fuel restrict radioactive releases to the environment. Intact cladding provides effective containment of radioactive material through all phases of spent fuel management. Assemblies that are free of dispersible fuel particulates can be retrieved after extended storage with low risk of environmental releases. Thus, oxidation of spent fuel to U_3O_8 powder during dry storage should be prevented in the event that the fuel is retrieved for reprocessing or repackaging.

Oxidizing atmospheres include air or an inert gas with significant quantities of oxidizing constituents. Current plans are to store spent fuel in inert cover gases. However, uncanistered fuel integrity should be maintained in case there is a loss of cover gas or in case radiolysis of water generates sufficient oxygen to permit the oxidation reaction to occur. It is desirable to license dry storage of spent fuel at the maximum temperature where the temperature-time exposure of the spent fuel is still maintained below the cumulative exposure required for cladding degradation by oxidation. Monitoring for inert cover gas integrity can be minimized or eliminated by maintaining storage temperatures below those where oxidation would occur. Storage facility cooling requirements are reduced by identifying the maximum acceptable temperature. Thus, the cost burden associated with dry storage is reduced. This paper describes an experimental program implemented by the DOE/PNL Commercial Spent Fuel Management Program to define the maximum temperatures for dry storage of spent fuel.

NONIRRADIATED FUEL DATA SUMMARY

Oxidation tests with nonirradiated UO_2 pellets were conducted to identify environmental and fuel characteristics that influence the oxidation rate. This information is summarized in Table I. Subsequent tests with spent fuel demonstrated significant differences in oxidation behavior between nonirradiated pellets and spent fuel. Significant weight gains in spent fuel specimens were observed without any U_3O_8 powder formation. Nonirradiated pellets formed U_3O_8 powder sooner with less total weight gain. Specimen weight gains are evidence that oxidation is occurring. Thus, additional tests with spent fuel are required to develop the final dry storage criteria. The results of the nonirradiated pellet tests were used to screen the significant variables and provide a focus for the spent fuel oxidation tests.

SPENT FUEL OXIDATION TESTS IN AIR

Spent fuel oxidation tests on Point Beach pressurized water reactor (PWR) spent fuel fragments (29,000 MWd/MTU burnup) were completed in atmospheric air at 230°C and 200°C. The fuel was heated in two identical air convection ovens. The ovens were located in gamma fields of 150,000 R/h and 600 R/h, respectively. The test specimens were periodically removed from the ovens, weighed, and visually examined for evidence of oxidation. Less frequently, the specimens were transferred to another facility for close-up examinations with a periscope and to remove small samples for x-ray diffraction analyses.

Results from these air tests are presented in Figs. 1 and 2. The slow initial rate of weight change (actually weight losses) has been attributed to drying of the test specimens. The oxidation behavior of the spent fuel did not correlate directly with that of nonirradiated pellets, even after correction was made for geometric surface area/volume ratios. Thus, further tests with spent fuel were required.

Periodic visual examinations were conducted to determine when U_3O_8 powder formation began. This point had been readily identified on the nonirradiated pellets after approximately 0.1 wt% total weight gain. In fact, powder formation on the nonirradiated pellets used as control specimens for the spent fuel tests was observed after 550 h in the high gamma field and after 965 h in the low gamma field at 230°C (Fig. 3). However, powder formation was not detected on the spent fuel fragments (Fig. 4), although significant (>3 wt%) weight gains had been observed. X-ray analysis of the spent fuel specimens after 965 h at 230°C and 1422 h at 200°C revealed weak, broad lines that are characteristic of an oxygen-rich tetragonal UO_2 phase. Even though the O/U ratio was 2.32, the diffraction pattern

TABLE I

Impacts of Oxidation Studies with Nonirradiated UO_2 on Spent Fuel Projects

Variable	Nonirradiated UO_2 Results	Guidance for Spent Fuel Studies
Temperature	Oxidation proceeds rapidly above 230°C.	Focus spent fuel studies below 230°C.
Aging	Aging in ambient air reduces oxidation rate.	Examine effect of aging.
Surface Area	Fragmentation increases oxidation rate approximately 5 to 7 times and has minor effect on particulate induction time.	Measure fragment size distribution.
Gamma Field	Gamma field enhances oxidation.	Impose gamma field spent fuel tests.
NO_2 in Cover Gas	Simulated NO_2 radiolysis product enhances oxidation.	Impose gamma field spent fuel tests.
Gadolinia	Gadolinia delays formation of U_3O_8 and results in the formation of fine fragments.	Include gadolinia spent fuel in tests; provide insight on fission product effects.
Density	Minor effect for closed porosity above 94.5% TD.	Measure density of spent fuel specimens.
Moisture	Moisture slightly increases oxidation rate.	Perform spent fuel tests under typical dry storage moisture conditions.
Temperature Changes	Oxidation response to temperature changes supports cumulative damage models.	Perform temperature change tests on spent fuel.
Thermal Cycles	Minor effect from thermal cycles.	Confirm slight thermal cycle effect for spent fuel.

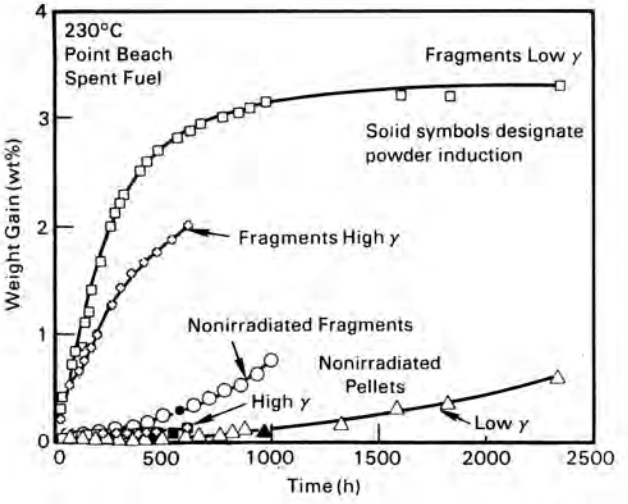

Fig. 1. Comparison of Oxidation of Spent Fuel with Nonirradiated UO_2 Pellets and Fragments at 230°C.

was not characteristic of U_3O_7. These results are consistent with the observation that no U_3O_8 powder had formed. There was no evidence of an underlying substrate.

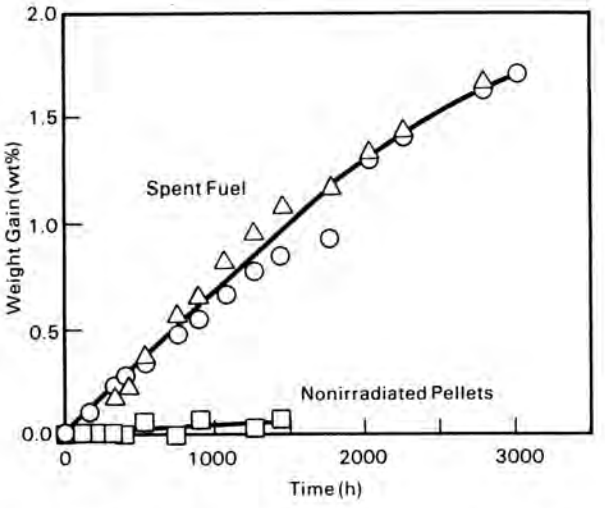

Fig. 2. Oxidation Behavior of Point Beach PWR Spent Fuel at 200°C (29,000 MWd/MTU burnup).

After 2337 h at 230°C and 3000 h at 200°C, spent fuel powder appeared to form. Closer inspection through a periscope revealed fragments and not a fine powder, with fragment sizes ranging up to 1 mm (Fig. 5). By this time the nonirradiated pellets had completely oxidized. X-ray analyses of these fragments yielded diffraction patterns with broad, very weak lines. Significant U_3O_7 formation was observed but no U_3O_8 was detected.

These tests were continued for an additional 2500 h. Additional x-ray diffraction samples were taken for subsequent analyses. These data will be reported at a later time.

Fig. 3. Unirradiated Pellets with U_3O_8 Formation.

Fig. 4. Point Beach Spent Fuel After 965 h at 230°C Prior to Formation of Fine Fragments.

CONTROLLED NITROGEN TEST SUMMARY

An evaluation of moist nitrogen as an inert cover gas has been completed at 275°C. Four test specimens were used: 1) Point Beach fuel fragments (29,000 MWd/MTU); 2) H. B. Robinson fuel fragments (30,000 MWd/MTU); 3) Shippingport fuel fragments (24,000 MWd/MTU); and 4) nonirradiated pellets. The integrity of the 20°C dew point nitrogen cover gas was verified with mass spectrometer analysis prior to the test. The oven was periodically shut down, a post-test gas sample was taken, and the specimens were weighed. The results of these tests are summarized in Table II. To date none of the fuel samples have shown any evidence of oxidation. These data have been used to support cask storage of spent fuel at 250°C in nitrogen.

ADDITIONAL SPENT FUEL TESTS

A significant data base is required to support licensing of dry spent fuel storage. As summarized in Table I, tests with nonirradiated UO_2 pellets demonstrated that various atmospheric contaminants (i.e., moisture, NO_x) could affect oxidation rates. Thus, a

Fig. 5. Fragmented Point Beach Spent Fuel.

decision was made to complete all future tests in controlled atmospheres. It was also decided to simultaneously complete a matrix of oxidation tests to generate the most useful data in the least time. Two arrays of six ovens each were fabricated (Fig. 6) that can be evacuated to 29 in. of Hg and will also maintain up to 5 psig of pressure. Each test oven is evacuated and backfilled to 2 psig with the desired cover gas. Pressure readings are continuously recorded. A leak can be readily detected and the system can be shut down before the test specimens are damaged. Operating temperatures of 100 to 400°C are

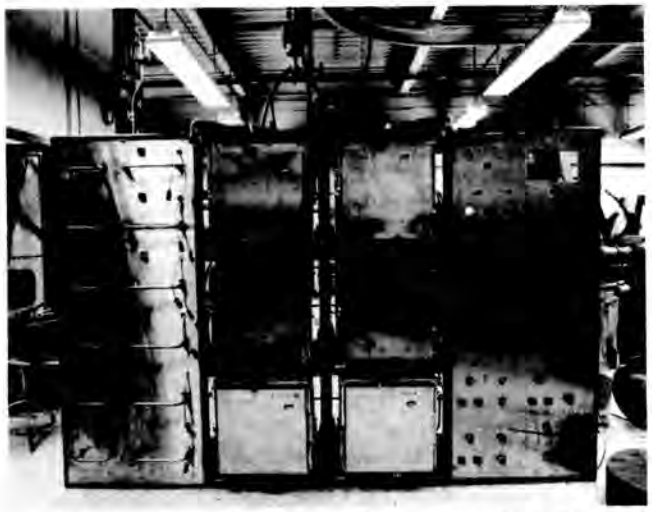

Fig. 6. Controlled Atmosphere Spent Fuel Test Oven Assembly.

possible with air or inert cover gases. Each oven can house up to 12 fuel specimens. The specimens can be periodically removed from the oven and weighed to ±0.002 grams on an electronic balance. One array of ovens is located adjacent to four cesium-137 sources for an enhanced gamma field. The other array is located across the hot cell from these sources where the gamma field is several orders of magnitude lower. Table III presents the test matrix that is in the process of being initiated. The effects of temperature, moisture, and gamma radiation on oxidation rate will be investigated. A range of different fuel types will be used to investigate the effects of burnup, cooling history, and fuel type (BWR versus PWR).

TABLE II

Results of Tests with Spent LWR Fuel in 275°C Moist Nitrogen Cover Gas

Specimen Number[a]	Time (h)	Weight (g)	Weight Change (g)
3A	0	18.416	0
3B	0	16.013	0
3C	0	29.763	0
3D	0	30.705	0
3A	432	18.413	-0.003
3B	432	16.01	-0.003
3C	432	29.756	-0.007
3D	432	30.703	-0.002
3A	1365	18.414	-0.002
3B	1365	16.01	0.003
3C	1365	29.758	-0.005
3D	1365	30.707	0.002

(a) 3A - H. B. Robinson spent fuel
3B - nonirradiated pellets
3C - Point Beach spent fuel
3D - Shippingport spent fuel.

TABLE III

Spent Fuel Oxidation Test Matrix

Temperature (°C)	Cover Gas	Dew Point (°C)
Low Gamma Field		
275	Nitrogen	20
400	Nitrogen	20
200	Air	20
180	Air	20
150	Air	20
150	Air	40
135	Air	20
High Gamma Field		
400	Nitrogen	20
180	Air	-40
180	Air	20
180	Air	40
165	Air	20
150	Air	20

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