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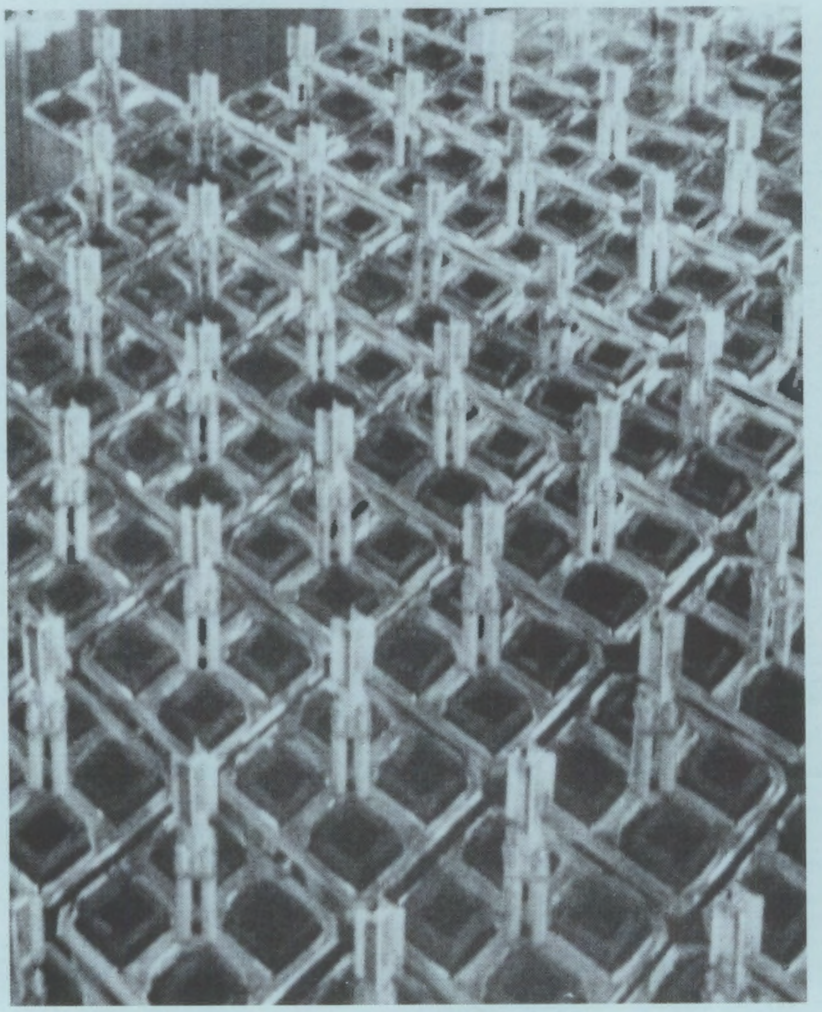
Assessment of Nitrogen as an Atmosphere for Dry Storage of Spent LWR Fuel

September 1985



Prepared for the U.S. Department of Energy
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Pacific Northwest Laboratory
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ASSESSMENT OF NITROGEN AS AN ATMOSPHERE FOR DRY STORAGE OF SPENT LWR FUEL

E. R. Gilbert
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September 1985

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the U.S. Department of Energy
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ABSTRACT

Interim dry storage of spent light-water reactor (LWR) fuel is being developed as a licensed technology in the United States. Because it is anticipated that license agreements will specify dry storage atmospheres, the behavior of spent LWR fuel in a nitrogen atmosphere during dry storage was investigated. In particular, the thermodynamics of reaction of nitrogen compounds (expected to form in the cover gas during dry storage) and residual impurities (such as moisture and oxygen) with Zircaloy cladding and with spent fuel at sites of cladding breaches were examined. The kinetics of reaction were not considered because it was assumed that the 20 to 40 years of interim dry storage would be sufficient for reactions to proceed to completion. The primary thermodynamic reactants were found to be NO_2, N_2O, H_2O_2, and O_2. The evaluation revealed that the limited inventories of these reactants produced by the source terms in hermetically sealed dry storage systems would be too low to cause significant spent fuel degradation. Furthermore, the oxidation of spent fuel to degrading O/U ratios is unlikely because the oxidation potential in moist nitrogen limits O/U ratios to values less than $UO_{2.006}$ (the equilibrium stoichiometric form in equilibrium with moist nitrogen). Tests were performed with bare spent UO_2 fuel and nonirradiated UO_2 pellets (with no Zircaloy cladding) in a nitrogen atmosphere containing moisture concentrations greater than encountered under dry storage conditions. These tests were performed for at least 1100 h at temperatures as high as 380°C, where oxidation reactions proceed in a matter of minutes. No visible degradation was detected, and weight changes were negligible.

SUMMARY

Nitrogen is being considered as an atmosphere for dry storage of spent light-water reactor (LWR) fuel in the United States because of its containability, abundance, thermal properties, and probable low reactivity with spent fuel and storage system components. Although dry storage of spent LWR fuel is not yet licensed in the United States, there is extensive favorable international experience with shipping spent fuel in nitrogen atmospheres. The objective of this evaluation is to determine if reactions between Zircaloy cladding and a nitrogen storage atmosphere and between exposed UO_2 at cladding breach sites and a nitrogen storage atmosphere could limit the application of nitrogen for dry storage. The effects of impurities (including residual oxygen, moisture from cask loading operations, moisture released from waterlogged fuel, and radiolysis products) were evaluated.

The thermodynamics for the reactions of zirconium and oxides of uranium with nitrogen compounds that form under conditions of radiolysis were assessed. The assessment showed that oxides of uranium will not react with nitrogen or residual moisture at expected dry storage temperatures. However, oxidizing impurities such as NO_2 and H_2O_2 formed by radiolysis of N_2 and H_2O or residual O_2 in a nitrogen cover gas system are reactive with UO_2. The extent of the degradation that could be caused by these impurities depends on their concentrations, the kinetics of the reactions, and the temperature-time exposure. The potential for moist nitrogen to oxidize UO_2 was shown to be limited to a stoichiometry corresponding to an O/U ratio not greater than $UO_{2.006}$.

The kinetics of the reactions were not considered. Because of the 20- to 40-year interim storage periods under consideration, it was conservatively assumed that all thermodynamically favorable reactions will proceed to completion. The reactivity of the very large Zircaloy fuel cladding surface area with these impurities and the potentially small inventory of radiolysis products in a dry storage system limits the amount of fuel that could be oxidized at cladding breach sites to a negligibly small amount.

Tests involving both nonirradiated UO_2 pellets at 225°C, 250°C, and 275°C and spent UO_2 fuel fragments at 275°C and 380°C were conducted at Pacific

Northwest Laboratory. An external gamma radiation field was imposed on the test atmosphere and specimens to provide radiolysis. After 7 weeks of testing, there were negligible weight gains and no change in the physical appearance of the specimens. The low reactivity of the nitrogen atmosphere for the duration of the test was evident by comparison with test results in air under comparable conditions. Tests in air for less than 1 week resulted in significant weight gains and powder generation by formation of low-density oxides of uranium.

Thus, it has been shown that no significant degradation occurred to UO_2 fuel in nitrogen within 7 weeks at temperatures up to 380°C. If reactions between the UO_2 fuel and the limited reactants available in the nitrogen storage atmosphere are assumed to go to completion during 20 to 40 years of dry storage, no deleterious degradation of the UO_2 fuel would be expected. This conclusion assumes that acceptable cask leak rates do not significantly increase. No credit was taken for the large surface area of Zircaloy cladding that provides an alternate sink for gaseous reactants.

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INTRODUCTION

Nitrogen is being considered as an atmosphere and heat transfer medium for interim dry storage of spent nuclear fuel. Nitrogen has been used internationally as an atmosphere for dry shipments of spent fuel. It is less prone to leakage than helium; and it is less reactive with spent fuel than air, which can oxidize fuel exposed at cladding defects at sufficiently high temperatures. Based on results of dry storage tests and demonstrations in air with spent light-water reactor (LWR) fuel (White et al. 1983; Einziger and Cook 1983; Johnson et al. 1983b; Einziger and Strain 1985; Johnson et al. 1985), storage temperatures that exceed yet-to-be-determined limits will require an inert cover gas to prevent potential cladding degradation by fuel oxidation in fuel rods with cladding breaches.

Evaluations sponsored by the U.S. Department of Energy (DOE) Commercial Spent Fuel Management (CSFM) Program at Pacific Northwest Laboratory (PNL)(a) have led to a less-restrictive but still-conservative temperature guideline that would allow fuel temperatures as high as 380°C during dry storage of spent fuel in inert cover gases in the United States (Johnson and Gilbert 1983b; Gilbert, Johnson, and Bailey 1985). The selection of this limit was based on results of dry storage tests and demonstrations in the Federal Republic of Germany (FRG) (Kaspar et al. 1982; Fleisch, Einfeld, and Luhrmann 1982) and conservative criteria for prevention of cladding breaches during dry storage by creep rupture (Chin and Madsen 1983).

Inert cover gases that have been used in dry storage tests and demonstrations include argon, helium, and neon (Johnson and Gilbert 1983b). Although nitrogen is not an inert gas, it is a potential atmosphere for spent fuel storage. Nitrogen has a lower thermal conductivity than helium; its higher density enables higher heat transfer by convection than in helium under thermal gradients in spent fuel dry storage systems; and it is relatively inexpensive. Because of its larger mass, nitrogen is less susceptible to leakage through small orifices by Knudsen flow than lower mass atoms of helium or neon (Dushman

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(a) Operated for DOE by Battelle Memorial Institute under Contract DE-AC06-76RLO 1830.

and Lafferty 1962). The specific benefits of using nitrogen instead of helium will depend on a number of design and operational considerations that require specific evaluation. The objective of the work reported this document is to provide a technical evaluation of the suitability of nitrogen as an interim dry storage atmosphere from the perspective of compatibility with spent Zircaloy fuel cladding, exposed UO_2 at cladding breach sites, and storage system components.

Although nitrogen reacts with metallic uranium to form uranium-nitrogen compounds, it is not thermodynamically favorable for nitrogen to reduce uranium oxides.

This report assesses data on the thermodynamics of zirconium and uranium oxide reactions with nitrogen and spent fuel cover gas impurities (moisture and radiolysis products), reviews experience with storing spent LWR fuel in nitrogen, and presents results of tests performed at PNL with UO_2 pellets and spent LWR fuel fragments in moist nitrogen. The PNL tests were designed to determine the compatibility of nonirradiated UO_2 pellets with moist nitrogen in a 200,000-R/h gamma irradiation field at 225°C, 250°C, and 275°C. Parallel tests were conducted on irradiated LWR fuel in moist nitrogen at 275°C and 380°C. The results from these tests are presented in this report. These tests were performed to support the use of nitrogen as an atmosphere in the Cask Characterization Test with spent boiling water reactor (BWR) fuel at the General Electric (GE) Morris fuel storage facility in Illinois (Newman 1985) and other cooperative DOE/utility dry storage demonstrations (Daily 1984). A broader consideration was to provide supporting data for qualifying nitrogen as a cover gas for licensed dry storage of spent fuel in the United States.

REVIEW OF PUBLISHED RESULTS

The results of tests conducted with spent fuel in the FRG, at Battelle Columbus Laboratories (BCL), at the Nevada Test Site (NTS), and at GE Morris are summarized in this section.

RADIOLYSIS

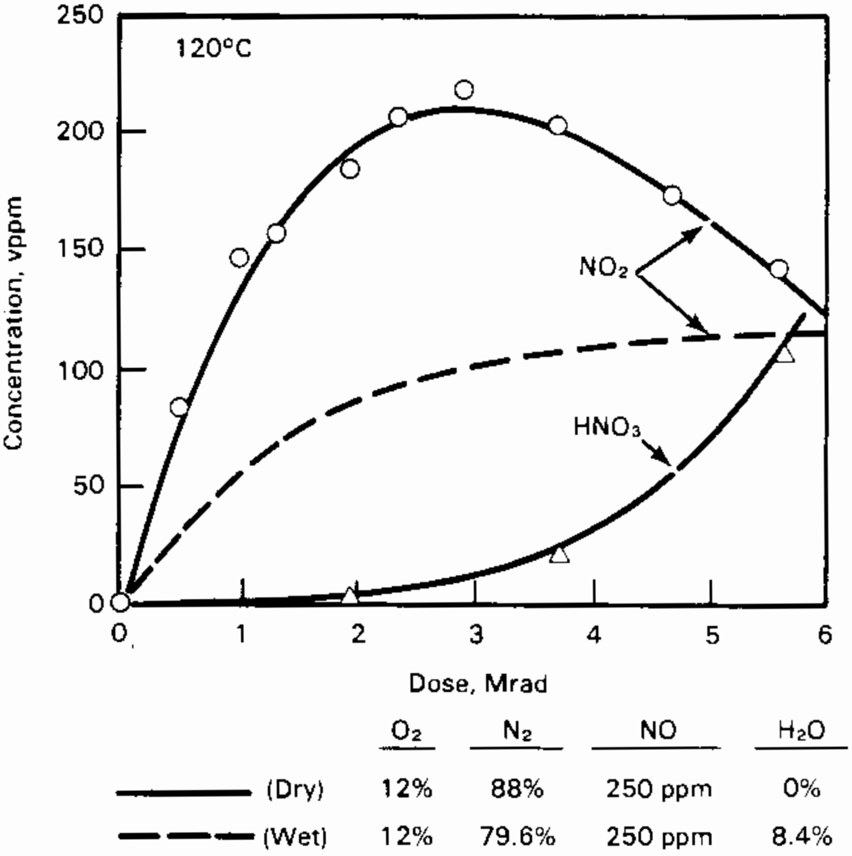
Some oxides of nitrogen are reactive with the oxides of uranium. Although oxides of nitrogen are not expected to occur naturally in significant quantities in nitrogen, they are formed by radiolysis of nitrogen and oxygen in a radiation field. Some N_xO_y compounds, such as NO_2, are strong oxidants and enhance oxidation rates in both reactive metals and ceramics (Samsonov 1973). Several of these compounds have negative free energies of reaction with UO_2 (Appendix D) and will oxidize UO_2 if the kinetics are sufficiently rapid. Radiolysis of nitrogen in the presence of oxygen or moisture in a radiation field generates N_xO_y compounds (Denaro and Jayson 1972). Radicals and products of radiolysis of nitrogen and moisture are shown in Table 1. Experimental results showing the effects of irradiation on the generation of NO_2 and HNO_3 in dry and moist nitrogen gases at 120°C are shown in Figure 1 (Tokunaga et al. 1978).

TESTS WITH SPENT FUEL AT FRG OBRIGHEIM

Results from tests performed by the FRG in moist nitrogen at the Obrigheim reactor have been reported for spent PWR fuel assemblies with fuel rods having burnup levels of 33,000 to 43,700 MWd/MTU (Peehs, Kuhnel, and Kaspar 1982;

TABLE 1. Radicals and Compounds Formed by Radiolysis of Moist Nitrogen

$$\begin{array}{ccccc}
\underline{\text{Gases}} & & \underline{\text{Reactive Species}} & & \underline{\text{Reaction Products}} \\
\left.\begin{array}{l} N_2 \\ O_2 \\ H_2O \\ \end{array}\right\} & \xrightarrow{\gamma} & \left\{\begin{array}{l} N_2^+,\ N^+,\ O_2^+,\ O^+,\ H_2O^+,\ OH^+, \\ H^+,\ H_2^+,\ N_2O^+,\ NO^+,\ O_3^+,\ H_3O^+, \\ O_3^-,\ O_2^-,\ N_2O^-,\ NO_2^-,\ N^*,\ O^*, \\ H_2^*,\ OH^*,\ NO^*,\ O_3^* \end{array}\right\} & \rightarrow & \left\{\begin{array}{l} N_2O_5,\ N_2O,\ NO_3, \\ NO_2,\ NO,\ HNO_2, \\ HNO_3,\ H_2O_2,\ O_3, \\ H_2 \end{array}\right.
\end{array}$$



	O_2	N_2	NO	H_2O
———— (Dry)	12%	88%	250 ppm	0%
— — — (Wet)	12%	79.6%	250 ppm	8.4%

FIGURE 1. NO_2 and HNO_3 Concentrations as a Function of Dose (1 Mrad = 50 days at 10^5 R/h) (Tokunaga et al. 1978)

Kaspar et al. 1982). The tests were conducted in a nitrogen-filled chamber immersed in and exposed to pool water for up to 60 days with a starting fuel temperature of 300°C and for up to 1 year with a starting fuel temperature of 400°C. No fuel assembly degradation was found during post-test visual examination. However, there were no known breached fuel rods in the test. Both fission gas analysis of the nitrogen atmosphere and sipping of the fuel rods were used to verify the absence of failed fuel.

Because the UO_2 fuel was protected by the Zircaloy-4 cladding, this test cannot be used to confirm the compatibility of spent UO_2 fuel with moist nitrogen (White et al. 1983; Johnson et al. 1983a; Kaspar et al. 1982; Peehs, Kuhnel, and Kaspar 1982). However, the Obrigheim test does indicate that there is no severe reaction between the ZrO_2-coated Zircaloy cladding and the moist nitrogen and is consistent with the low rates of reaction of Zircaloy with oxygen and nitrogen at 325°C (Johnson and Gilbert 1983a). However, a quantitative post-test examination of the Zircaloy cladding was not reported.

TESTS WITH SPENT FUEL AT BATTELLE COLUMBUS LABORATORIES

The tendency for reactor-breached spent fuel rods to contain moisture was the basis for an ultrasonic method used to identify breached fuel rods (Cruickshank 1985; Boehm and Foerch 1985). In one series of spent fuel examinations, 47 fuel rods from 36 fuel assemblies were found to contain moisture (Walton et al. 1985). To determine the behavior of water-logged spent fuel in dry storage, in-air tests on breached BWR spent fuel rods were sponsored by the DOE CSFM Program and conducted by BCL. The results from these tests provide supporting information on the compatibility between UO_2 or Zircaloy cladding and nitrogen (Johnson et al. 1983b; Kohli et al. 1985).

The tests were continued for 2100 h at 325°C. The oxygen in the air cover gas was depleted within approximately 10 h by oxidation of the spent fuel through reactor-induced cladding breaches. The residual nitrogen was repeatedly evacuated and replaced with air. During the 10-h period while the oxygen was being depleted, the pressure decreased by approximately 20% and then became stable. During this stable pressure period, the cover gas was composed primarily of nitrogen. If the spent fuel or the Zircaloy cladding were reacting with the nitrogen atmosphere, a corresponding pressure reduction would be expected. The only pressure change that was measured following depletion of the oxygen was a slow increase during the first few hundred hours of the test. This slow pressure increase was attributed to approximately 4 g of bound moisture that was slowly released from the water-logged fuel and not to any reaction between the nitrogen atmosphere and the spent fuel that would have caused a pressure reduction. After a few hundred hours, the moisture release was too slow to generate a measurable pressure increase.

The reactor-breached spent fuel test in air and a parallel test in argon showed that up to 10 g of moisture per rod was released (Johnson et al. 1983a; Kohli et al. 1985). Over half of this moisture was released very rapidly under vacuum and therefore would have been released during vacuum drying operations in a storage cask. A small amount of moisture (up to 4 g per rod) appeared to be bound in the fuel and was slowly released (Kohli et al. 1985). Other studies indicate that some moisture may be bound in the fuel.(a)

TESTS WITH SPENT FUEL AT NTS-EMAD

Significant amounts of N_2O (up to 2500 vppm) were found in samples of the air atmosphere surrounding a single PWR spent fuel assembly in the Fuel Temperature Test (FTT) initiated at 275°C at the Engine Maintenance and Disassembly Facility (EMAD) at NTS (Johnson et al. 1985). The N_2O buildup was attributed to radiolysis of nitrogen and oxygen by gamma irradiation of the air atmosphere surrounding the spent fuel assembly and subsequent reactions between nitrogen and oxygen. Post-test visual examination and swipe tests for smearable fuel contamination of the fuel assembly revealed no evidence for reaction between the atmosphere and the Zircaloy cladding or the fuel at an unidentified cladding breach that formed within the first 2 months of the 2-year test.

TESTS WITH SPENT FUEL AT GE MORRIS

A BWR Cask Characterization Test was performed at the away-from-reactor spent fuel storage pool at GE Morris in Illinois (Newman 1985). The primary objective of the test was to verify heat transfer and radiation shielding code predictions and cask performance. Nitrogen was used as the atmosphere for one series of tests with up to 52 BWR spent fuel assemblies enclosed in the cask. A sketch of a typical dry storage cask is shown in Figure 2; the cask is approximately 4.9 m high with an internal cavity diameter of about 155 cm. If all of the pressure associated with cask evacuation to 1 torr prior to filling the cask with nitrogen were due to moisture, the dew point would be approximately -20°C (4 g of moisture in the 5.4×10^6 cm^3 cask atmosphere). Analyses of gas samples taken from the cask atmosphere indicated a total moisture content of 0.6 to 2.7 g. Post-test visual examination and pre- and post-test wet sipping of the fuel showed no detectable degradation to the spent fuel assemblies.

(a) Olander, Sherman, and Balooch 1982; Beyer and Hann 1977; Spalaris and Magerth 1963; Denovan, Ashley, and Longhurst 1971; Ferrari 1963.

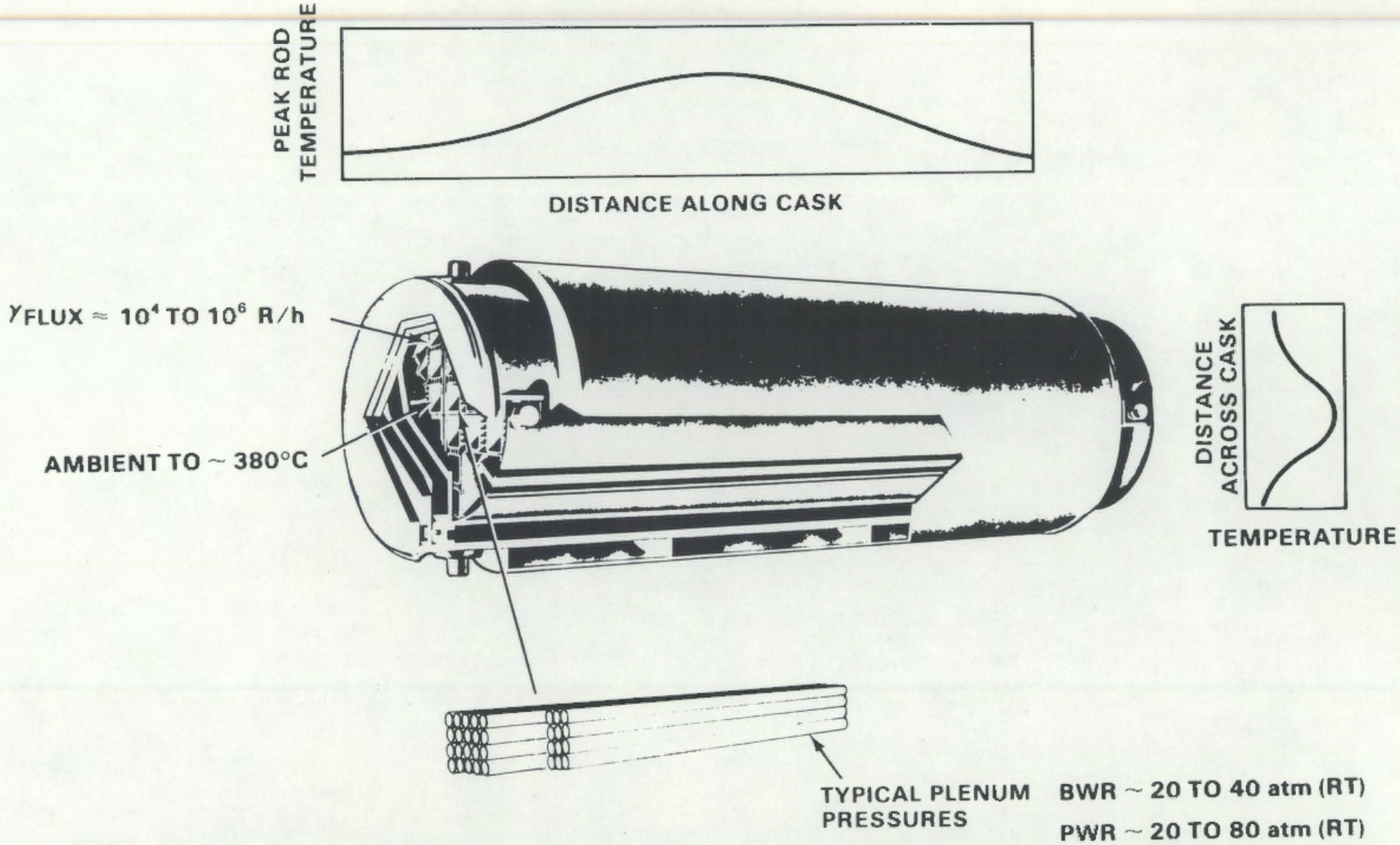


FIGURE 2. Typical Cask Used for the BWR Cask Characterization Test

DESCRIPTION OF PNL FUEL EXPERIMENTS

DOE/CSFM-sponsored tests were conducted on both nonirradiated and irradiated UO_2 to determine if the radiolysis products of moist nitrogen would oxidize UO_2. The experimental procedures for UO_2 pellet and spent fuel tests at PNL have been published (White et al. 1983; Knox, Gilbert, and White 1985). Bare fuel pellets (approximately 5.5 g each) or bare spent fuel fragments (3 to 5 mm edge lengths) were placed in ovens with a controlled atmosphere of: 1) air-1 vol% NO_2, 2) moist air or moist nitrogen in a gamma field generated by a cobalt-60 source, or 3) moist nitrogen in a gamma field generated by CsCl capsules. The air-1 vol% NO_2 mixture was used to synthesize a radiolysis product, while gamma radiation was used to simulate radiolytic effects of the gamma field generated by spent fuel in a dry storage installation. Gas samples taken from the test atmospheres were analyzed before and after the test. The tests were periodically interrupted for weighing and visual examination of specimens.

The objectives of the tests performed in air were to determine if the presence of a synthesized radiolysis product would affect the oxidation of UO_2 and to determine if radiolysis of air would affect the oxidation of UO_2. The fuel gained weight with increasing time at temperature due to the oxidation. Oxides of uranium between UO_2 and UO_3 are shown in the phase diagram in Figure 3.

PRELIMINARY TESTING IN AIR-1 vol% NO_2

Tests simulating the effect of radiolysis products on oxidation of UO_2 in air were performed at PNL by using air containing 1 vol% NO_2. The rate of weight gain in low-density nonirradiated UO pellets (Figures 4 and 5) (White et al. 1983) was significantly higher than in air. The significantly larger weight gains for one of the pellet tests described in Figure 4 for air-1 vol% NO_2 was attributed to its low theoretical density of 95%. The other pellets tested in air-1 vol% NO_2 and air had approximately 96% theoretical density.

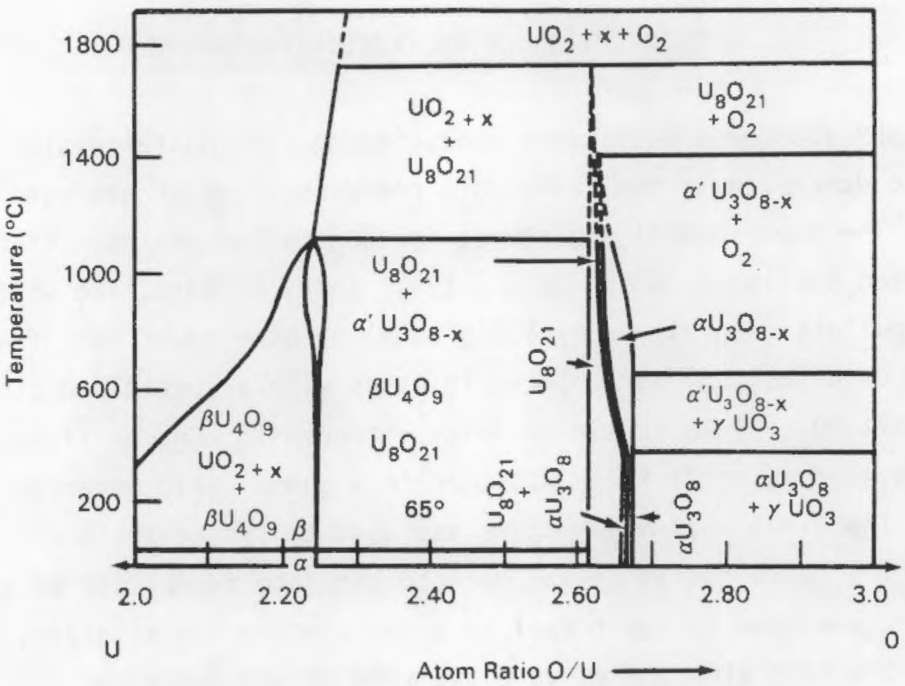


FIGURE 3. U-O Phase Diagram (Roth et al. 1981)

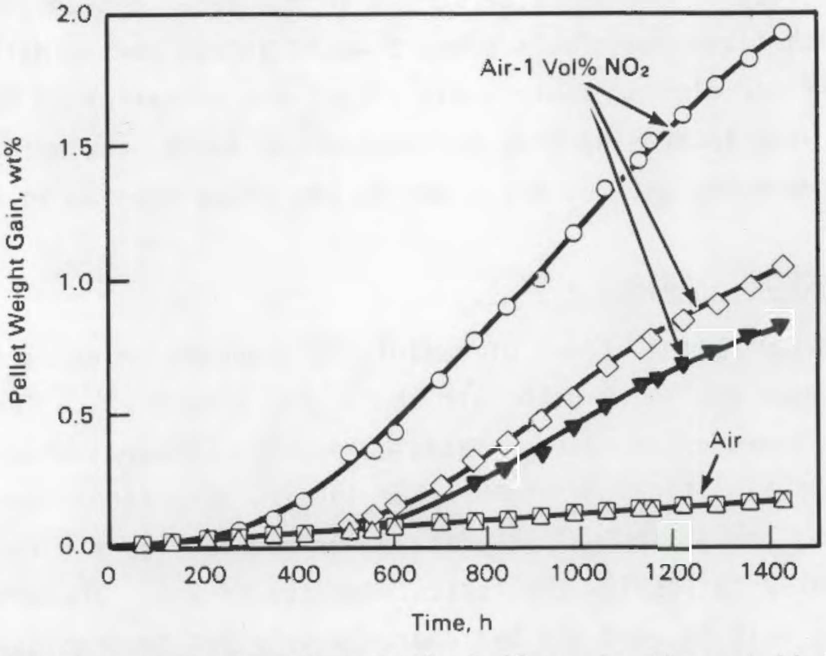


FIGURE 4. Oxidation of Nonirradiated UO_2 Pellets in Air and Air-1 vol% NO_2 at 215°C. The large weight increases for the highest curve are attributed to its low density of only 95% theoretical compared with the approximately 96% theoretical for the other pellets.

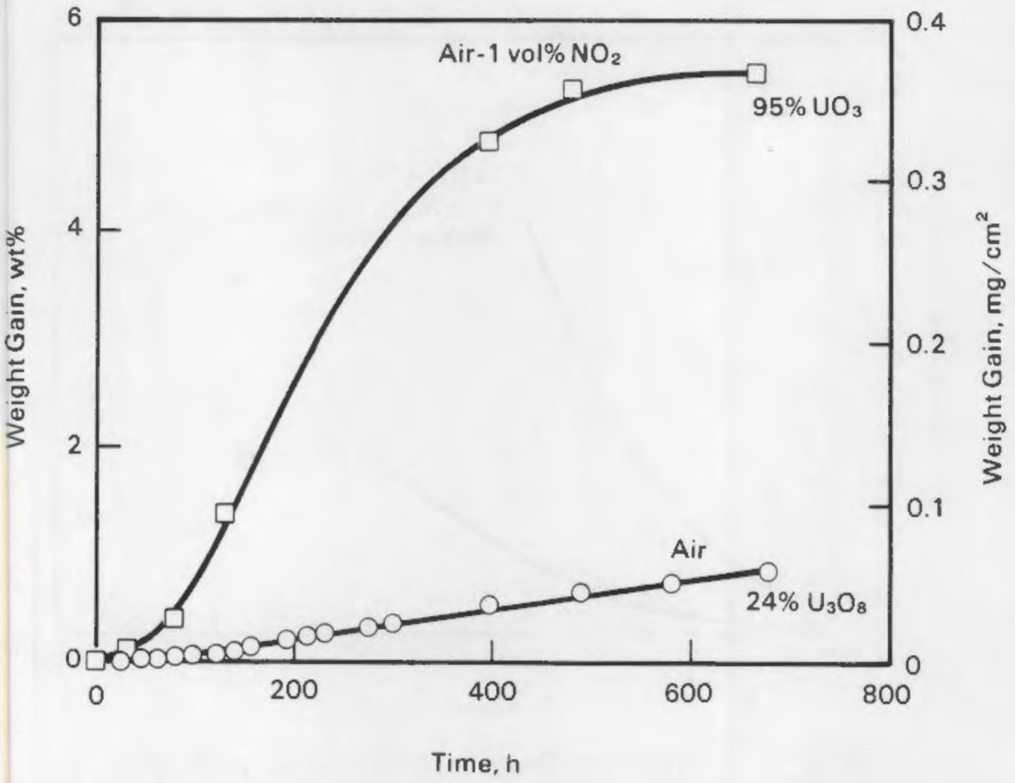


FIGURE 5. Oxidation of Nonirradiated UO_2 Pellets in Air and Air-1 vol% NO_2 at 250°C

PRELIMINARY TESTING WITH A GAMMA FIELD IN AIR

Tests were performed at PNL to explore the effects of a gamma field in producing reactive radiolysis products and the subsequent effects on UO_2 oxidation. These tests, conducted in a 200,000 R/h gamma field generated by a cobalt-60 source, resulted in accelerated oxidation of nonirradiated UO_2 pellets (Figure 6). The final product was UO_3 powder. Although no N_2O was found in chemical analyses of test atmosphere gas samples at the completion of the test, the concentration of N_2O in the test chamber was slightly greater than 100 vppm after less than 1 week of irradiation. These tests demonstrated that radiolysis products form in a gamma field of intensity typical of that expected in a dry storage installation containing spent LWR fuel and that UO_2 pellet oxidation was accelerated in a gamma field typical of a dry storage installation.

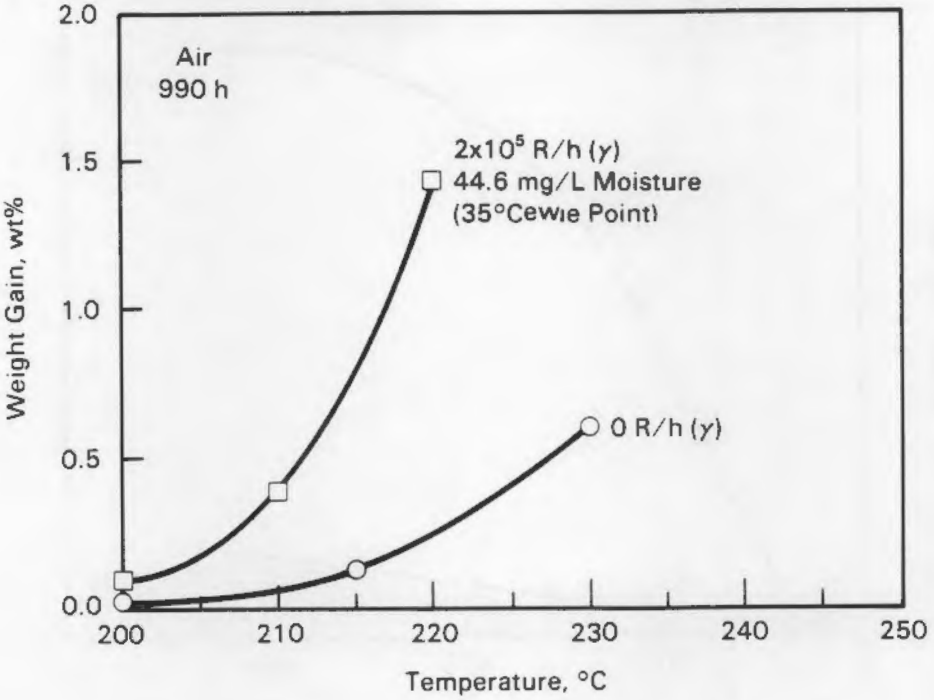


FIGURE 6. Effect of Gamma Radiation on Oxidation of Nonirradiated UO_2 Pellets

NONIRRADIATED BARE UO_2 PELLET TESTS IN NITROGEN

A nonirradiated UO_2 pellet was placed in each of three gas-tight capsules that were evacuated and backfilled with nitrogen at a 20°C dew point. The three cells were maintained at 225°C, 250°C, and 275°C, respectively, in a gamma field of 200,000 R/h. Pellet densities were 96.4%, 95.1%, and 95.7% of theoretical, respectively, at each temperature. The oxygen concentration measured in pre- and post-test samples of the atmosphere was less than 0.01 vol%. The gamma field intensity is typical of the gamma field generated by a 35,000 MWd/MTU spent PWR fuel assembly approximately 7 years after reactor discharge or a 20,000 MWd/MTU spent BWR fuel assembly approximately 3 years after reactor discharge (Croff 1980). The pellet appearance was observed, and the weight change was determined after 455 and 1165 cumulative hours at the test temperature. As shown in Table 2, there were no measurable weight changes. The reduced nitrogen concentration shown in the gas analyses in Table 2 is associated with the formation of CO_2. The pellets retained a glossy surface

TABLE 2. Results of Tests on Nonirradiated UO_2 Pellets in Moist Nitrogen in a Gamma Field of 200,000 R/h

Temperature, °C	Time, h	Weight, g	Weight Change, g	CO_2, vol%	H_2, vol%	N_2, vol%
225	0	5.4432	--	<0.01	<0.01	99.9
250	0	5.3199	--	<0.01	<0.01	99.9
275	0	5.2688	--	<0.01	<0.01	99.9
225	445	5.4431	-0.0001	0.28	0.1	99.6
250	445	5.3197	-0.0002	0.63	<0.01	99.4
275	295	5.2687	-0.0001	0.51	<0.01	99.5
225	1165	5.4430	-0.0002	0.18	0.08	99.74
250	1165	5.3196	-0.0003	0.46	0.02	99.43
275	1015	5.2689	0.0001	0.3	0.06	99.64

with no evidence of cracking or degradation. The specimens and reaction products were analyzed by x-ray diffraction to determine if changes in composition and crystal form had resulted from exposure to the moist nitrogen cover gas; no changes were found.

The gas volume of each test cell was 163 cm^3. The nitrogen cover gas was introduced into the cell at a dew point of 20°C or a moisture content of 0.017 mg/cm^3, which corresponds to 2.8 mg of moisture in each test cell. The amount of oxygen combined in 2.8 mg moisture is approximately 2.5 mg. Based on the changes in pellet weights shown in Table 2, no measurable oxidation occurred. Most pellet tests in air showed visible U_3O_8 formation at a weight increase of 2 to 8 mg per UO_2 pellet. Therefore, it is possible that if all of the oxygen reacted with the fuel pellet U_3O_8 formation might have been observed. However, based on the lack of weight increase, it appears that there was no significant oxidation. It can be concluded that either the radiolysis was inadequate to produce oxygen for reaction with the UO_2 or the oxygen was not available for reaction.

During preliminary tests for verification of procedures and equipment integrity, oxygen inleakage caused U_3O_8 powder to form within 1 week after initiation of a test at 275°C. In subsequent tests after the leaks were eliminated and only moist nitrogen was introduced, no weight gain was found and no powder formed. From the contrast in pellet behavior in air and in nitrogen, it was apparent that the nitrogen cover gas did not result in significant pellet degradation.

BARE SPENT FUEL FRAGMENT TESTS IN NITROGEN

Tests on bare spent fuel fragments were conducted at 275°C with fuel from Point Beach (29,000 MWd/MTU peak burnup), H. B. Robinson (33,000 MWd/MTU peak burnup), and Shippingport (34,000 MWd/MTU peak burnup; 24,000 MWd/MTU average burnup) reactors in moist (20°C dew point) nitrogen. Tests were also conducted at 380°C with six spent fuel samples at a dew point of 30°C. Since the dew point in a dry storage atmosphere is expected to be on the order of -40 to -20°C, these tests were performed at a higher moisture level than spent fuel will be subjected to in storage. The formation of radiolytic products increases with moisture content (Figure 1). Therefore, the atmospheres in these tests should be more reactive than those formed in a dry storage installation. The pre- and post-test specimen weights and results of gas analyses are shown in Tables 3 and 4.

A cesium chloride gamma irradiation source was placed next to the test ovens to simulate the gamma field associated with spent fuel during dry storage. Gamma fields of approximately 12,000 R/h were calculated for the center of the 275°C oven, and gamma fields of 120,000 R/h were measured for the 380°C oven. The 16,000-cm^3 volume in the test oven contained 276 mg of moisture at a 20°C dew point. The amount of oxygen combined with this moisture (0.17 moles O_2) is sufficient to oxidize approximately 13% of the approximately 95 g of spent fuel in the 275°C oven or approximately 7% of the approximately 170 g of spent fuel in the 380°C oven to U_3O_8. The specimens were periodically weighed to monitor oxidation and visually inspected for evidence of reaction (color change and powder formation). As shown in Tables 3 and 4, weight changes were small or not measurable. There were no visible changes in appearance of the spent fuel fragments or the nonirradiated UO_2 pellets. Spent fuel is very reactive with oxygen at 380°C and U_3O_8 forms in less than 1 h at 360°C (Einziger and Strain 1985). The differences in weight changes 1 h and 24 h after cooling at ambient temperature are attributed to moisture pickup by the UO_2 during exposure to the hot cell atmosphere.

TABLE 3. Results of Tests with Spent LWR Fuel in 275°C Moist Nitrogen Cover Gas (20°C dew point)

Specimen[(a)]	Time, h	Weight, g	Weight Change, g	CO_2, mol%	H_2, mol%	N_2, mol%
3A	0	18.416	0	<0.01	<0.01	99.9
3B	0	16.013	0	<0.01	<0.01	99.9
3C	0	29.863	0	<0.01	<0.01	99.9
3D	0	30.705	0	<0.01	<0.01	99.9
3A	432	18.413	-0.003	0.22	<0.01	99.6
3B	432	16.010	-0.003	0.22	<0.01	99.6
3C	432	29.756	-0.007	0.22	<0.01	99.6
3D	432	30.703	-0.002	0.22	<0.01	99.6
3A	1365	18.414	-0.002	0.29	0.09	99.4
3B	1365	16.010	-0.003	0.29	0.09	99.4
3C	1365	29.758	-0.005	0.29	0.09	99.4
3D	1365	30.707	0.002	0.29	0.09	99.4

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

The bare fuel test results show a significant accumulation of CO_2 in the test atmosphere. The CO_2 was apparently released from the spent fuel during heating. The lack of reaction between the fuel and the CO_2 is consistent with the positive free energy of reaction shown in Appendix C.

TABLE 4. Results of Tests with Spent LWR Fuel in 380°C Moist Nitrogen Cover Gas (31°C dew point)

Specimen(a)	Time, h	Weight, g	Weight Change, g	CO_2, mol%	H_2, mol%	N_2, mol%	O_2, mol%	CH_4, mol%
5-11	0	33.287	0	<0.01	<0.01	99.97	0.03	<0.01
5-12	0	23.668	0	<0.01	<0.01	99.97	0.03	<0.01
5-13	0	28.435	0	<0.01	<0.01	99.97	0.03	<0.01
5-14	0	30.026	0	<0.01	<0.01	99.97	0.03	<0.01
5-15	0	22.629	0	<0.01	<0.01	99.97	0.03	<0.01
5-16	0	29.277	0	<0.01	<0.01	99.97	0.03	<0.01
5-11	505(b)	32.894	-0.393	0.77	0.34	98.17	0.04	0.36
5-12	505	23.281	-0.387	0.77	0.34	98.17	0.04	0.36
5-13	505	28.054	-0.381	0.77	0.34	98.17	0.04	0.36
5-14	505	30.030	0.004	0.77	0.34	98.17	0.04	0.36
5-15	505	22.296	-0.333	0.77	0.34	98.17	0.04	0.36
5-16	505	29.276	0.001	0.77	0.34	98.17	0.04	0.36
5-11	505(c)	33.294	0.007	0.77	0.34	98.17	0.04	0.36
5-12	505	23.684	0.016	0.77	0.34	98.17	0.04	0.36
5-13	505	28.432	-0.003	0.77	0.34	98.17	0.04	0.36
5-14	505	30.032	0.006	0.77	0.34	98.17	0.04	0.36
5-15	505	22.632	0.003	0.77	0.34	98.17	0.04	0.36
5-16	505	29.278	0.001	0.77	0.34	98.17	0.04	0.36
5-11	1104(b)	33.296	0.009	0.59	0.16	98.45	0.02	0.38
5-12	1104	23.686	0.018	0.59	0.16	98.45	0.02	0.38
5-13	1104	28.445	0.010	0.59	0.16	98.45	0.02	0.38
5-14	1104	30.035	0.009	0.59	0.16	98.45	0.02	0.38
5-15	1104	22.638	0.009	0.59	0.16	98.45	0.02	0.38
5-16	1104	29.284	0.007	0.59	0.16	98.45	0.02	0.38

(a) 5-11 - H. B. Robinson bare spent fuel fragments (N-9B Section L4)
5-12 - H. B. Robinson bare spent fuel fragments (N-9B section L3)
5-13 - Point Beach bare spent fuel fragments ("A" section 5-13, 63 cm above bottom end)
5-14 - Point Beach bare spent fuel fragments ("A" section 5-14, 58 cm above bottom end)
5-15 - Shippingport bare spent fuel fragments ("B" section 5-15 end piece)
5-16 - Shippingport bare spent fuel fragments ("B" section 6-16 end piece).

(b) 1 to 4 h after cooling at ambient temperature.

(c) 24 h after cooling at ambient temperature.

DISCUSSION

The effects of moisture in breached fuel rods on cladding integrity and spent fuel canisters have been evaluated (Woodley 1984). This report focuses on the thermodynamics of reactions between Zircaloy cladding and reactive constituents and between fuel at a cladding breach with the quantities of reactive constituents that could affect the spent fuel storage in nitrogen during 20 to 40 years of interim dry storage.

THERMODYNAMICS OF URANIUM-OXYGEN COMPOUNDS

Numerous studies have reported on the thermodynamics and phase diagrams for uranium-oxygen compounds.(a) The O/U ratio for UO_2 increases in an oxidizing environment with temperature-dependent rates. Key chemical reactions are presented in Appendix A. These chemical reactions are characterized by negative free energies of reaction, indicating that they are thermodynamically favorable. All of these reactions have been observed in the temperature regimes proposed for dry storage of spent LWR fuel. U_3O_7 and U_4O_9 have theoretical densities similar to or slightly greater than that for UO_2 (11.4 g/cm^3 for U_3O_7 versus 10.97 g/cm^3 for UO_2). Consequently, the oxidation products for those compounds do not lead to cladding degradation. However, U_3O_8 and UO_3 have lower theoretical densities than UO_2 (8.35 g/cm^3 for U_3O_8 and 7.9 g/cm^3 for UO_3). Sufficient formation of U_3O_8 at the site of a breach in an LWR fuel rod can produce permanent deformation of the cladding as well as crack formation and extension. Tests with bare spent fuel result in spalling of surface grains with high O/U ratios along grain boundaries, while the interior of the pellet grains remain at lower ratios of O/U because of slow solid state diffusion.

Since irradiated UO_2 contains numerous fission products as impurities, its oxidation behavior may be significantly different from that of nonirradiated UO_2 and may depend on burnup (Gilbert, White, and Knox 1985). Approximately

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(a) Levin et al. 1964; Levin and McMurdie 1975; Roth et al. 1981; Samsonov 1973; Barin and Knacke 1973; Barin, Knacke, and Kubaschewski 1977; Stull and Prophet 1971; Wick and Block 1963; Wagman et al. 1982.

2 at.% fissions of the fission products depend on the burnup and operating temperature of the fuel. The amount of fuel oxidation occurring during dry storage will impact the water solubility of the fuel and fuel performance during subsequent disposal in a repository.

THERMODYNAMICS OF REACTIONS BETWEEN URANIUM OXIDE AND WATER

Reactions between the oxides of uranium and moisture have positive free energies of formation and are energetically unfavorable during dry storage (Appendix C). Therefore, the oxides of uranium would not react with moisture in a dry storage atmosphere where radiolysis of moisture is not a significant factor.

Radiolysis of moisture produces H_2O_2 as well as H_2 and O_2 (Denaro and Jayson 1972). The reactions of H_2O_2 and O_2 with the oxides of uranium are energetically favorable in expected dry storage temperature ranges (Appendices A and E). Even though H_2O is not reactive with the oxides of uranium (Appendix C), the formation of H_2O_2 and O_2 would provide a potentially degrading oxidation reaction with spent LWR fuel at cladding breach sites, if concentrations were sufficiently high.

THERMODYNAMICS OF REACTIONS BETWEEN OXIDES OF URANIUM AND N_xO_y

The free energies for the reactions of the oxides of uranium with N_xO_y indicate that no reaction is expected with pure nitrogen (Appendix D). An evaluation was performed to determine the maximum acceptable operating temperature of UO_2 in flowing nitrogen (Berry 1971; Levy and Foster 1961). The temperature limit was predicted to be 1200°C. The criteria for deriving this limit are not clear since formation of UN from UO_2 is associated with a positive free energy of formation of 157 kcal/mole of UO_2 at 1200°C (Samsonov 1973).

The free energies of reaction for NO_2, NO, and N_2O with UO_2 are negative, indicating that reactions are favorable. The presence of NO_2 or a gamma field accelerated oxidation of UO_2 pellets in air (Figures 4, 5, and 6). However, no reaction between moist nitrogen and either UO_2 pellets or spent fuel fragments could be detected at temperatures from 225 to 380°C (Tables 2, 3, and 4).

THERMODYNAMICS OF REACTIONS BETWEEN ZIRCONIUM AND NITROGEN COVER GAS

The free energies of reaction for zirconium with O_2, H_2O_2, N_2O, and N_2 are negative; thus, the reactions are energetically favorable (Appendix F). However, the external spent fuel cladding surface is coated with ZrO_2 that was formed during fuel manufacture or during exposure to reactor coolant prior to discharge (Johnson and Gilbert 1983a). Reaction of nitrogen with the protective ZrO_2 coating is shown in Appendix F to be thermodynamically unfavorable. Consequently, little reaction is expected between nitrogen and the spent fuel cladding during dry storage. Accelerated oxidation of Zircaloy has been attributed to radiolytic-formed H_2O_2 in aqueous environments (Johnson 1977). However, this reaction is expected to be of little consequence in dry storage since the increase in ZrO_2 thickness due to the reaction of the small inventory of H_2O_2 with the nearly 4 million cm^2 of Zircaloy cladding surface area in a typical dry storage cask is negligible.

APPLICATION OF RESULTS TO INTERIM DRY STORAGE OF SPENT FUEL IN NITROGEN

Even though spent LWR fuel is not expected to react with nitrogen during dry storage, impurities and compounds formed by radiolysis of moisture contaminants are potentially reactive. The primary sources of water are:

- residual moisture remaining in the storage system after incomplete drying from pool loading operations
- release from water-logged reactor-breached fuel rods
- desorption of moisture from crud on the surface of fuel rods
- outgassing of storage component surfaces.

Results from the BWR Cask Characterization Test showed that the residual moisture remaining from pool loading operations, desorption of moisture from crud, and outgassing of storage component surfaces is less than 3 g. The tests with reactor-breached whole rods at BCL indicated that the amount of moisture available from water-logged reactor-breached fuel rods after cask vacuum drying is less than 4 g per breached rod. Therefore, a dry storage cask containing 52 BWR 7x7 spent fuel assemblies with 0.1% failed fuel rods (Garzarolli, von Jan, and Stehle 1979) would contain approximately three breached fuel rods (<12 g moisture) or a total moisture content of <16 g after vacuum drying. The oxygen combined in this moisture is approximately 14 g or 0.4 moles of oxygen and could convert approximately 60 UO_2 pellets to U_3O_8 if all of the oxygen reacted with bare fuel at cladding breach sites. It is considerably more likely that the moisture would react with the Zircaloy cladding to produce a negligible increase in ZrO_2 thickness.

The data base reviewed includes international shipping of spent fuel in nitrogen (shipping involving cask storage times up to several months), FRG and U.S. storage tests and demonstrations in moist nitrogen, and tests with bare fuel in moist nitrogen. None of these results indicated any reaction between spent fuel assemblies or bare fuel with nitrogen. The temperatures were sufficiently high that significant reactions would have resulted with the bare fuel in air. For example, during a test with spent fuel in nitrogen at 380°C, air was inadvertently allowed to enter the oven. The spent fuel specimens were converted to a fine powder in less than 10 min.

The lack of reaction between UO_2 and moist nitrogen may seem surprising since the free energy of reaction of UO_2 with radiolysis products such as N_2O and NO_2 indicate that reactions are thermodynamically favorable. However, the oxidation potential of UO_2 is reduced in the presence of moist nitrogen.

The oxygen potential of pure water vapor at pressure po_{H_2O} is calculated as shown below (calculations provided by D. R. Olander, University of California).

Assuming water equilibrium according to:

$$H_2(g) + 1/2\ O_2(g) = H_2O(g) \tag{1}$$

yields

$$K = \frac{p_{H_2O}}{(p_{H_2})(p_{O_2})^{1/2}} = \exp\ (-\Delta Go/RT) \tag{2}$$

where p_i is the partial pressure of species i in the equilibrium gas and Go is the standard free energy of formation of water vapor:

$$\Delta Go = -250.8 + 57.72(T/10^3)\text{ kJ/mole} \tag{3}$$

At 200°C, Go = -223.5 kJ/mole (54.1 kcal/mole).

Neglecting the reaction of water with Zircaloy cladding or other components of the dry storage installation to calculate the maximum oxygen potential of the gas, material balances on oxygen and hydrogen yield:

$$po_{H_2O} = p_{H_2O} + 2p_{O_2} \tag{4}$$

$$po_{H_2O} = p_{H_2O} + p_{H_2} \tag{5}$$

Equations (2), (4), and (5) can be solved for the oxygen partial pressure:

$$po_{H_2O} = 2K(p_{O_2})^{3/2} + 2p_{O_2} \tag{6}$$

Since the last term can be neglected because K is very large,

$$p_{O_2} = (po_{H_2O}/2K)^{2/3} \tag{7}$$

the oxygen potential of the moist nitrogen is:

$$\Delta G_{O_2} = RT \ln p_{O_2} \tag{8}$$

The above simplified analysis provides the maximum oxygen potential of the moist nitrogen in the absence of radiolysis effects since it neglects the reaction with the Zircaloy cladding. Performing the above calculation for 200°C gives -39 kcal/mole for the oxygen potential of moist nitrogen. According to the data listed for the first reaction in Appendix A, the oxygen potential of the two-phase UO_2-U_4O_9 mixture is 0.5 x -32 kcal/mole = -16 kcal/mole. Since this value is larger than that calculated for moist nitrogen, moist nitrogen cannot thermochemically oxidize UO_2 to U_4O_9. The equilibrium fuel stoichiometry based on an oxygen potential of -39 kcal/mole at 200°C is $UO_{2.006}$ (Olander 1976). Consequently, reaction between the oxidants and the nearly 4 million cm^2 of more accessible Zircaloy cladding would be more likely.

Even though the quantity of reactants in a spent fuel storage installation containing nitrogen is not sufficient to cause significant degradation to the fuel assemblies, the effect of additional reactants introduced by inleakage of air should be considered. Storage casks are leak tested to a rate of not greater than 1 x 10^{-6} cm^3/s helium (GNS 1983). The corresponding leak rate for oxygen by Knudson flow (Dushman and Lafferty 1962) is approximately 3 x 10^{-7} cm^3/s. At this rate, less than 0.2 L or 0.01 moles of oxygen would leak into the cask in 20 years. If this oxygen reacted with fuel at the site of a single cladding breach, no more than two fuel pellets would be oxidized to U_3O_8. Since this oxygen would be dispersed and would not react with fuel as a single breach site (the Zircaloy cladding offers approximately 4 million cm^2 of reaction surface), the oxygen inleakage cannot be expected to produce significant fuel degradation.

CONCLUSIONS

The following conclusions are made:

- A thermodynamic assessment of the compatibility of Zircaloy cladding and UO_2 with pure nitrogen indicates that no chemical reactions are predicted during interim dry storage.
- Although Zircaloy cladding can thermodynamically react with impurity oxidants, moisture, and products of radiolysis, cladding degradation is expected to be negligible in the dry storage temperature regime. The small inventory of reactants plus the negligible oxygen inleakage cannot produce significant reaction layer thickness with the nearly 4 million cm^2 of Zircaloy surface area in a BWR storage cask containing 52 spent fuel assemblies.
- Based on thermodynamic considerations, chemical reactions are possible between impurity oxidants (O_2 and radiolysis products such as H_2O_2 and NO_2) in the storage system cover gas and UO_2 exposed at cladding defects. However, the oxygen potential in moist nitrogen is in equilibrium with a fuel stoichiometry of $UO_{2.006}$; and, therefore, low-density powders with stoichiometry of $UO_{2.67}$ and above cannot form. Most oxidizing reactants would react with the very large, more reactive, accessible Zircaloy cladding surface. Furthermore, the quantity of reactants expected in a dry storage system are insufficient to produce significant fuel degradation.
- Only a small amount of experimental data on the stability of UO_2 in nitrogen is available. The limited experience dealing with the exposure of Zircaloy-clad fuel with nitrogen indicated that nitrogen is nonreactive with uranium oxide fuel, Zircaloy, and the oxide film on Zircaloy cladding.
- Tests on spent fuel fragments in moist nitrogen with a superimposed gamma irradiation field showed negligible weight gains and no changes in the physical appearance of the fuel specimens for up to 7 weeks at 275°C and 380°C.

- Tests with nonirradiated UO_2 pellets in moist nitrogen and with a superimposed gamma irradiation field showed no measurable weight gain or change in the physical appearance of the UO_2 pellets for times up to 7 weeks at 225°C, 250°C, and 275°C.

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APPENDIX A

THERMODYNAMICS OF CHEMICAL REACTIONS OF OXIDES OF URANIUM WITH OXYGEN AT 200°C

APPENDIX A

THERMODYNAMICS OF CHEMICAL REACTIONS OF OXIDES OF URANIUM WITH OXYGEN AT 200°C

Reactions and Free Energies of Reactants and Products, kcal/mole	Free Energy of Reaction,(a) kcal/mole
$4UO_2 + 1/2\ O_2 = U_4O_9$ -1076 -12 -1120	-32 (Barin and Knacke 1973)
$3UO_2 + 1/2\ O_2 = U_3O_7$ -740 0 -775	-35(b) (Wagman et al. 1982)
$3UO_2 + O_2 = U_3O_8$ -807 -24 -890	-59 (Barin and Knacke 1973)
$3UO_2 + O_2 = U_3O_8$ -718 0 -775	-57 (Samsonov 1973)
$U_3O_7 + 1/2\ O_2 = U_3O_8$ -775 0 -805	-30(b) (Wagman et al. 1982)
$U_3O_8 + 1/2\ O_2 = 3UO_3$ -890 -12 -915	-13 (Barin and Knacke 1973)
$U_3O_8 + 1/2\ O_2 = 3UO_3$ -775 0 -789	-14 (Samsonov 1973)

(a) A negative free energy of reaction indicates the reaction is thermodynamically favorable.

(b) 25°C temperature.

APPENDIX B

CONCENTRATIONS OF FISSION PRODUCTS FROM 35,000 MWd/MTU PWR FUEL AT DISCHARGE AND 10 YEARS AFTER DISCHARGE

APPENDIX B

CONCENTRATIONS OF FISSION PRODUCTS FROM 35,000 MWd/MTU PWR FUEL AT DISCHARGE AND 10 YEARS AFTER DISCHARGE

(Croff 1980)

Element	Concentration, wt%	
	At Discharge	After 10 yr
H	5.99×10^{-6}	3.41×10^{-6}
Zn	2.3×10^{-9}	0
Ga	1.26×10^{-9}	0
Ge	3.87×10^{-5}	3.86×10^{-5}
As	9.09×10^{-6}	8.84×10^{-6}
Se	5.49×10^{-3}	5.49×10^{-3}
Br	1.64×10^{-3}	1.64×10^{-3}
Kr	3.98×10^{-2}	3.83×10^{-2}
Rb	3.53×10^{-2}	3.67×10^{-2}
Sr	9.76×10^{-2}	8.22×10^{-2}
Y	5.09×10^{-2}	4.95×10^{-2}
Zr	3.90×10^{-1}	4.00×10^{-1}
Nb	3.75×10^{-3}	4.17×10^{-7}
Mo	3.57×10^{-1}	3.67×10^{-1}
Tc	8.88×10^{-2}	8.92×10^{-2}
Ru	2.51×10^{-1}	2.29×10^{-1}
Rh	3.76×10^{-2}	4.16×10^{-2}
Pd	1.32×10^{-1}	1.49×10^{-1}
Ag	6.48×10^{-3}	6.39×10^{-3}
Cd	8.49×10^{-3}	8.60×10^{-3}
In	1.36×10^{-4}	1.41×10^{-4}
Sn	5.52×10^{-3}	5.40×10^{-3}
Sb	1.88×10^{-3}	1.15×10^{-3}
Te	6.00×10^{-2}	6.00×10^{-2}
I	2.87×10^{-2}	2.82×10^{-2}
Xe	5.81×10^{-1}	5.81×10^{-1}
Cs	2.85×10^{-1}	2.41×10^{-1}
Ba	1.44×10^{-1}	1.87×10^{-1}
La	1.35×10^{-1}	1.34×10^{-1}
Ce	3.03×10^{-1}	2.63×10^{-1}
Pr	1.23×10^{-1}	1.27×10^{-1}
Nd	3.98×10^{-1}	4.36×10^{-1}
Pm	1.28×10^{-2}	9.28×10^{-4}
Sm	8.41×10^{-2}	9.62×10^{-2}
Eu	1.94×10^{-2}	1.72×10^{-2}
Gd	1.11×10^{-2}	1.38×10^{-2}
Tb	2.02×10^{-4}	1.90×10^{-4}
Dy	1.00×10^{-4}	1.12×10^{-4}
Ho	1.11×10^{-5}	1.11×10^{-5}
Er	3.12×10^{-6}	3.15×10^{-6}
He	2.07×10^{-5}	8.13×10^{-5}
Tl	1.19×10^{-16}	2.44×10^{-15}
Pb	2.6×10^{-11}	4.73×10^{-9}
Bi	1.16×10^{-14}	1.57×10^{-13}
Po	2.10×10^{-18}	1.46×10^{-16}
At	1.08×10^{-23}	3.63×10^{-24}
Rn	1.08×10^{-16}	2.20×10^{-15}
Fr	1.09×10^{-19}	1.09×10^{-19}
Ra	9.63×10^{-13}	1.81×10^{-11}
Ac	3.46×10^{-13}	2.85×10^{-12}
Th	4.47×10^{-8}	2.61×10^{-7}
Pa	1.33×10^{-8}	1.85×10^{-8}
U	9.55×10^{1}	9.55×10^{1}
Np	5.25×10^{-2}	4.53×10^{-2}
Pu	7.42×10^{-1}	7.18×10^{-1}
Am	1.21×10^{-2}	4.38×10^{-2}
Cm	4.15×10^{-3}	2.27×10^{-3}
Bk	1.58×10^{-10}	5.01×10^{-14}
Cf	7.19×10^{-11}	2.06×10^{-10}
Es	1.30×10^{-14}	0

Total grams from fission products = 3.70×10^{4} g/tU

Total grams from actinides = 9.63×10^{5} g/tU

1.00×10^{6} g/tU

APPENDIX C

THERMODYNAMICS OF CHEMICAL REACTIONS BETWEEN OXIDES OF URANIUM AND H_2O, CO, AND CO_2 AT 200°C

APPENDIX C

THERMODYNAMICS OF CHEMICAL REACTIONS BETWEEN OXIDES OF URANIUM AND H_2O, CO and CO_2 AT 200°C

Reactions and Free Energies of Reactants and Products, kcal/mole	Free Energy of Reaction,(a) kcal/mole
$4UO_2 + H_2O = U_4O_9 + H_2$ -1076 -80 -1120 -15	21 (Barin and Knacke 1973)
$3UO_2 + H_2O = U_3O_7 + H_2$ -740 -55 -775 0	20(b) (Wagman et al. 1982)
$3UO_2 + 2H_2O = U_3O_8 + 2H_2$ -807 -160 -889 -30	48 (Barin and Knacke 1973)
$UO_2 + H_2O = UO_3 + H_2$ -269 -80 -306 -15	28 (Barin and Knacke 1973)
$3U_4O_9 + 5H_2O = 4U_3O_8 + 5H_2$ -3360 -400 -3556 -75	129 (Barin and Knacke 1973)
$U_3O_7 + H_2O = U_3O_8 + H_2$ -775 -55 -805 0	25(b) (Wagman et al. 1982))
$U_3O_8 + H_2O = 3UO_3 + H_2$ -805 -55 -819 0	41(b) (Wagman et al. 1982)
$UO_3 + H_2O = UO_3 \cdot H_2O$ -306 -80 -384	2 (Baron and Knacke 1973; Barin, Knacke, and Kubaschewski 1977)
$UO_3 + H_2O = UO_3 \cdot H_2O$ -273 -55 -333	-5(b) (Wagman et al. 1982)
$UO_3 + 2H_2O = UO_3 \cdot 2H_2O$ -273 -110 -390	-7(b) (Wagman et al. 1982)
$3UO_2 + 2CO = U_3O_8 + 2C$ -807 -96 -889 308	322 (Barin and Knacke 1973)
$3UO_2 + CO_2 = U_3O_8 + C$ -807 -118 -889 154	190 (Barin and Knacke 1973)

(a) A negative free energy of reaction indicates the reaction is thermodynamically favorable.

(b) 25°C temperature.

APPENDIX D

THERMODYNAMICS OF CHEMICAL REACTIONS BETWEEN OXIDES OF URANIUM AND N_xO_y AT 200°C

APPENDIX D

THERMODYNAMICS OF CHEMICAL REACTIONS BETWEEN OXIDES OF URANIUM AND N_xO_y AT 200°C

Reactions and Free Energies of Reactants and Products, kcal/mole	Free Energy of Reaction,(a) kcal/mole
$\underset{-269}{UO_2} + \underset{-11}{1/2\ N_2} = \underset{-78}{UN} + \underset{-24}{O_2}$	178 (Barin and Knacke 1973)
$\underset{-269}{UO_2} + \underset{-10}{1/2\ NO_2} = \underset{-306}{UO_3} + \underset{-6}{1/4\ N_2}$	-33 (Barin and Knacke 1973)
$\underset{-269}{UO_2} + \underset{-3}{NO} = \underset{-306}{UO_3} + \underset{-11}{1/2\ N_2}$	-45 (Barin and Knacke 1973)
$\underset{-269}{UO_2} + \underset{-6}{N_2O} = \underset{-306}{UO_3} + \underset{-22}{N_2}$	-53 (Barin and Knacke 1973)
$\underset{-807}{3UO_2} + \underset{-20}{NO_2} = \underset{-889}{U_3O_8} + \underset{-11}{1/2\ N_2}$	-73 (Barin and Knacke 1973)
$\underset{-807}{3UO_2} + \underset{-6}{2NO} = \underset{-889}{U_3O_8} + \underset{-22}{N_2}$	-98 (Barin and Knacke 1973)
$\underset{-807}{3UO_2} + \underset{-12}{2N_2O} = \underset{-889}{U_3O_8} + \underset{-44}{2N_2}$	-114 (Barin and Knacke 1973)
$\underset{-807}{3UO_2} + \underset{-17}{1/2\ N_2O_4} = \underset{-889}{U_3O_8} + \underset{-11}{1/2\ N_2}$	-76 (Barin and Knacke 1973)
$\underset{-807}{3UO_2} + \underset{-6}{2NO} = \underset{-889}{U_3O_8} + \underset{-22}{N_2}$	-98 (Barin and Knacke 1973)

(a) A negative free energy of reaction indicates the reaction is thermodynamically favorable.

APPENDIX E

THERMODYNAMICS OF CHEMICAL REACTIONS BETWEEN H_2O_2 AND OXIDES OF URANIUM AT 200°C

APPENDIX E

THERMODYNAMICS OF CHEMICAL REACTIONS BETWEEN H_2O_2 AND OXIDES OF URANIUM AT 200°C

Reactions and Free Energies of Reactants and Products, kcal/mole	Free Energy of Reaction,(a) kcal/mole
$3UO_2 + 2H_2O_2 = U_3O_8 + 2H_2O$ -807 -116 -889 -160	-126 (Barin and Knacke 1973)
$3U_4O_9 + 5H_2O_2 = 4U_3O_8 + 5H_2O$ -3360 -290 -3556 -400	-306 (Barin and Knacke 1973)
$U_3O_7 + H_2O_2 = U_3O_8 + H_2O$ -775 -25 -805 -55	-60(b) (Wagman et al. 1982)

(a) A negative free energy of reaction indicates the reaction is thermodynamically favorable.

(b) 25°C temperature.

APPENDIX F

THERMODYNAMICS OF REACTIONS OF ZIRCONIUM WITH O_2, H_2O_2, H_2O, N_2, N_2O, AND NO_2 AT 200°C

APPENDIX F

THERMODYNAMICS OF REACTIONS OF ZIRCONIUM WITH O_2, H_2O_2, H_2O, N_2, N_2O, AND NO_2 AT 200°C

Reactions and Free Energies of Reactants and Products, kcal/mole	Free Energy of Reaction,[a] kcal/mole
$Zr + O_2 = ZrO_2$ -5 -24 -269	-240 (Barin and Knacke 1973)
$Zr + 2H_2O_2 = ZrO_2 + 2H_2O$ -5 -116 -269 -160	-308 (Barin and Knacke 1973)
$Zr + 2H_2O = ZrO_2 + 2H_2$ -5 -16 -269 -30	-134 (Barin and Knacke 1973)
$Zr + 1/2\ N_2 = ZrN$ -5 -11 -92	-76 (Barin and Knacke 1973)
$Zr + 2NO = ZrO_2 + N_2$ -5 -6 -269 -22	-280 (Barin and Knacke 1973)
$Zr + 2N_2O = ZrO_2 + 2N_2$ -5 -12 -269 -44	-296 (Barin and Knacke 1973)
$Zr + NO_2 = ZrO_2 + 1/2\ N_2$ -5 -20 -269 -11	-255 (Barin and Knacke 1973)
$ZrO_2 + 1/2\ N_2 = ZrN + O_2$ -269 -22 -92 -24	175 (Barin and Knacke 1973)

(a) A negative free energy of reaction indicates the reaction is thermodynamically favorable.

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