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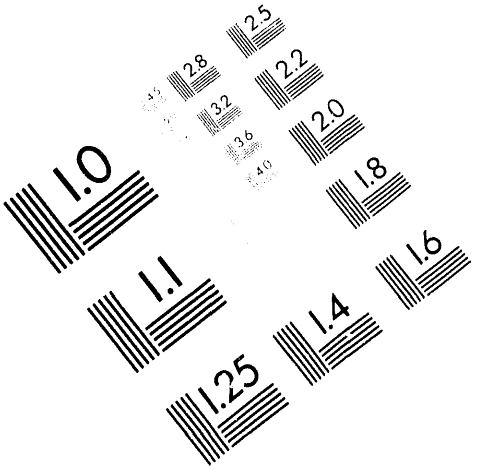

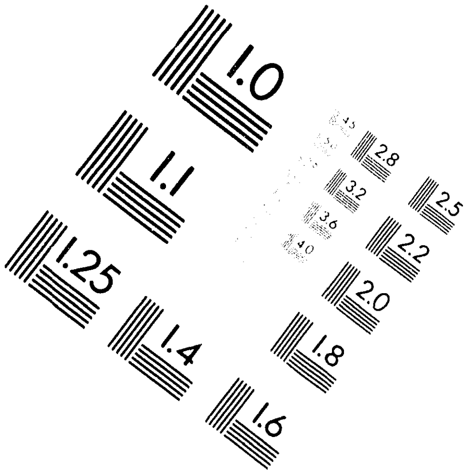

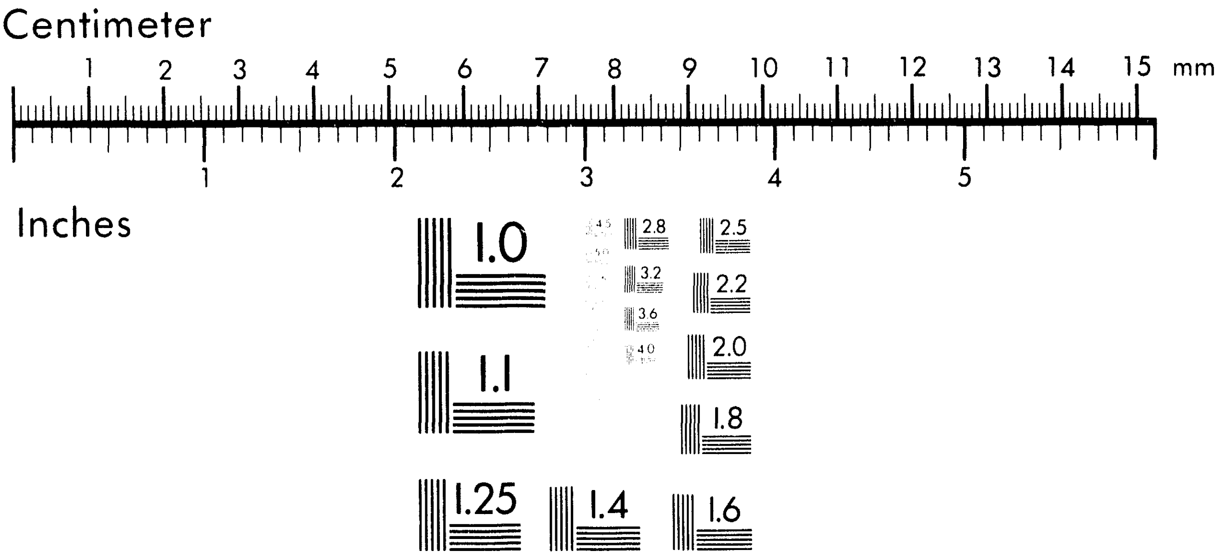

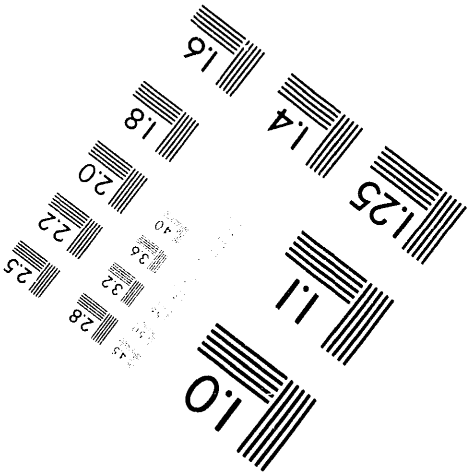

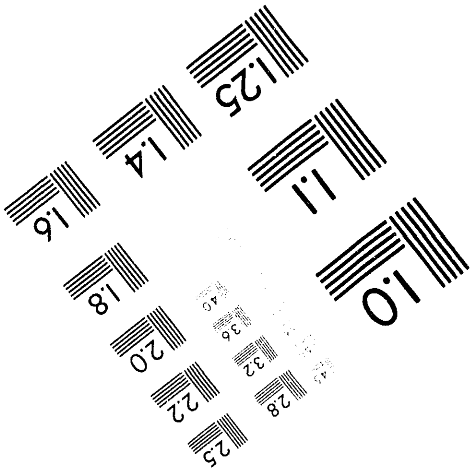

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Comparison of Uranium Dissolution Rates From Spent Fuel and Uranium Dioxide

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COMPARISON OF URANIUM DISSOLUTION RATES FROM SPENT FUEL AND URANIUM DIOXIDE

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ABSTRACT

Two similar sets of dissolution experiments, resulting from a statistical experimental design, were performed in order to examine systematically the effects of temperature (25-75°C), dissolved oxygen (0.002-0.2 atm overpressure), pH (8-10) and carbonate concentrations ($2\text{-}200\times10^{-4}$ molar) on aqueous dissolution of UO_2 and spent fuel. The average dissolution rate was 8.6 $mg/m^2\cdot day$ for UO_2 and 3.1 $mg/m^2\cdot day$ for spent fuel. This is considered to be an insignificant difference; thus, unirradiated UO_2 and irradiated spent fuel dissolved at about the same rate. Moreover, regression analyses indicated that the dissolution rates of UO_2 and spent fuel responded similarly to changes in pH, temperature, and carbonate concentration. However, the two materials responded very differently to dissolved oxygen concentration. Approximately half-order reaction rates with respect to oxygen concentration were found for UO_2 at all conditions tested. At room temperature, spent fuel dissolution (reaction) rates were nearly independent of oxygen concentration. At 75°C, reaction orders of 0.35 and 0.73 were observed for spent fuel, and there was some indication that the reaction order with respect to oxygen concentration might be dependent on pH and/or carbonate concentration as well as on temperature.

I. INTRODUCTION

The long term effects of the interactions between spent fuel, as a radioactive waste form, and groundwaters must be anticipated to safely dispose of spent fuel in an underground repository. Spent fuel dissolution and subsequent transport processes in groundwater are generally considered to be the main routes by which radionuclides could be released from a geological repository. Because uranium dioxide (UO_2) is the primary constituent of spent nuclear fuel, the dissolution of the UO_2 spent fuel matrix is regarded as a necessary first step for release of about 98% of the radioactive fission products contained within the UO_2 matrix. The intrinsic UO_2 dissolution rate sets an upper bound on the aqueous radionuclide release rate, even if the fuel is substantially degraded by other processes such as oxidation. If the fuel is substantially degraded to other oxidation states, then their dissolution responses also must be provided. The release rate is reduced for the solubility-limited actinides (U, Np, Pu and Am), which account for most of the long-lived radioactivity in spent fuel, when colloids are not present. At this point the outcome of the reactions of the spent nuclear fuel with the groundwater cannot be predicted, because a satisfactory release model based on observable chemical processes is not yet available.

There have been many investigations of the dissolution of UO_2, spent fuel and uraninite (a naturally occurring UO_2 mineral) in aqueous solutions, under both reducing and oxidizing conditions, and as a function of various other environmental variables. Several reviews have been written, most recently by Grambow.[1] Important variables considered in the investigations included pH, temperature, oxygen fugacity and carbonate/bicarbonate concentrations. The data vary due to the differences in experimental purpose and methods, the diverse history of the fuel samples, the formation of secondary phases during the tests, the complexity of the solution and the surface chemistry of UO_2, and the surface area measurements of the test specimens. The results of such diverse studies are difficult to compare and interpret. Several researchers have developed equations to correlate dissolution rates as a function of relevant variables. However, none of the rate laws is universal, and inconsistencies or incompatibilities among the proposed laws are common.

The purpose of the work reported here was to systematically determine the effect of temperature and important water chemistry variables on the dissolution rates of the UO_2 matrix phase in both unirradiated UO_2 and spent fuel. The UO_2 work was done at Lawrence Livermore National Laboratory (LLNL) and the spent fuel work was done at Pacific Northwest Laboratories (PNL). Unirradiated UO_2 represents fresh reactor fuel with no burnup. The two data sets comprise the first direct comparison of UO_2 and spent fuel dissolution rates and provide insight into the effect of fuel burnup.

The exact chemistry of groundwater in an underground repository is not certain, but groundwater has typical constituents, such as carbonates, sulfates, chlorides, silicates, and calcium. Water taken from wells near Yucca Mountain, contains all of these ions and has a pH near 8. Of the anions commonly found in groundwater, carbonate is considered to be the most aggressive towards UO_2 and, as such, is a conservative surrogate for all anions in groundwater. The data obtained from the tests described here can be used to: 1) identify important parameters that control the dissolution rates of the UO_2 matrix phase of spent fuel, 2) estimate bounding values for UO_2 and spent fuel matrix dissolution rates, and 3) develop a release model for radionuclides from spent fuel that will be used in waste package design and in performance assessment.

II. EXPERIMENTAL DESCRIPTION

Benefiting from the experiences of previous studies, the intrinsic dissolution rates of UO_2 and spent fuel were determined by using a single pass flow-through method that has been successfully used in the study of the dissolution kinetics of glass and other minerals[2], of UO_2[3,4] and of spent fuel.[5] The advantage of the single pass flow-through technique is that flow rates and specimen size can be controlled so that the UO_2 dissolves under conditions that are far from solution saturation (no precipitation of dissolved products). Under such conditions, the steady-state dissolution rates are directly proportional to the effective surface area of the specimen.[6] Thus, the dependence of UO_2 dissolution kinetics on pH, temperature, oxygen and carbonate/bicarbonate concentrations can be evaluated.

A set of experiments was selected to examine systematically the effects of temperature (25-75°C), dissolved oxygen (0.002-0.2 atm overpressure), pH (8-10) and carbonate concentrations (0.2-20 millimol/L) on UO_2 dissolution. The high temperature was limited to 75°C, because temperatures nearer to 100°C induce experimental difficulties in an aqueous, flowthrough system. Later extrapolation of results close to 100°C should not induce significant errors. The carbonate concentrations bracketed the typical groundwater concentration of 1 millimol/L. The oxygen pressure represented the atmospheric value and down two orders of magnitude to a minimally oxidizing atmosphere. The pH covered a value typical of groundwaters (pH=8) to very alkaline conditions.

Test solutions were prepared using analytical-reagent grade chemicals and deionized water. Each solution was continuously sparged with nitrogen or argon gas containing fixed concentrations of oxygen and carbon dioxide to maintain the desired dissolved oxygen concentration and pH of the solution. The test solutions flowed through the sample cells at rates between 5 and 25 mL/hr. One to three times per week, effluent from the cells was collected, acidified to prevent uranium adsorption on the sample vial walls, and analyzed for uranium content using a phosphorescence analyser. Dissolution rates were calculated from uranium concentrations multiplied by flow rates and divided by surface areas of the test specimens. After steady-state dissolution rates were achieved, the flow rates were occasionally changed to ensure that the observed dissolution rates remained unchanged. Dissolution rates will not be affected by changing flow rates, if the reaction is not solubility- or transport-limited.

The spent fuel used in the PNL tests was ATM-103, a PWR fuel with a burnup of 33 MWd/kgM and a fission gas release of 0.25%.[7] The UO_2 specimens used at LLNL were about 1 cm across and consisted of large crystallites containing dislocation substructures, i.e., low-angle grain boundaries. They are part of a very large batch produced and hand-picked in the early 1960's at PNL. Specimens from this batch were used at several laboratories to determine many of the reference physical properties of UO_2, such as melting point, thermal conductivity, hardness and electrical conductivity. The O/U ratio is 2.00 by coulometry after electrical conductivity measurements.[8]

III. TEST MATRIX

In order to test for nonlinear effects of the four variables on the uranium dissolution rate from UO_2 and spent fuel, experiments at three different values of each variable were required. The chosen settings were pH = 8, 9, 10, temperatures of 25°, 50° and 75°C, oxygen partial pressures of 0.002, 0.02 and 0.2 atm, and total carbonate concentrations of 0.2, 2 and 20 millimol/L. A statistical experimental design approach was used to select the experiments to be performed and to reduce the number of required experiments. A model including nonlinear effects and interactions of all four variables has at least 15 terms.

A classical three-level, full-factorial experimental design consists of the 81 (3^4) different, possible combinations of variable settings from the four variables at low, medium and high values. Performing such a large number of experiments on both UO_2 and spent fuel was unrealistic. A D-optimal design of 17 experiments was chosen using the RS/Discover computer program from BBN Software.[9] Seventeen experiments provides the minimum two additional degrees of freedom to perform a regression fit to a fifteen-term model with experimental data that has inherent error. One experiment at middle values of the four variables was performed in triplicate to test the reproducibility of the experiments. The 17 different experiments were selected from the 81 experiments in the full-factorial design. The D-optimal approach significantly reduced the number of experiments required by classic factorial or fractional-factorial designs. These experiments are uniformly distributed over the four-dimensional variable space. This set of 19 experiments, including the two replicates, allows us to fit a fifteen-term second-order model discussed earlier. The extra degrees of freedom permit tests for experimental variability. If smaller models are satisfactory, the full set of 19 dissolution conditions and rates provides for higher confidence in the models. The same test matrix of 19 experiments was run for both UO_2 and spent fuel, except for the three identical spent fuel tests at 20% oxygen..

In addition to replication experiments in the test matrix, one extra experiment was done in the spent fuel study (run 20 of Table 1). Three additional experiments not in the original UO_2 test matrix were performed (runs 24-26 of Table 1). The supplemental UO_2 experiments came from the seven remaining candidates at 75°C and 20% oxygen in the original 81-candidate set of the test matrix. These additional tests provided more data to examine the effect of oxygen concentrations on dissolution rates.

IV. RESULTS

The results of the combined uranium dioxide and spent fuel test matrices are given in Table 1. The data are presented in tabular and equation formats, because two-dimensional graphs cannot adequately display dissolution rates as a function of four independent variables.

Several approaches to dissolution modeling have been explored, including different model forms, as well as full and partial regressions that depend on some assumptions about the degree of variable dependency. The classical observed chemical kinetic rate law for homogeneous reactions is one of the models being examined and takes the following well-known general form:[10]

$$\text{Rate} = k[A]^a[B]^b[C]^c \ldots \exp(-E_a/RT), \qquad (1)$$

This generalized form of the rate law is for homogeneous gas or liquid reaction systems. It does not take into consideration the possibly complex liquid-solid reaction at the UO_2 or spent fuel surface. Additional term(s) are needed to account for this element of the reaction, and any radiation effects in the spent fuel, but they are unknown at this time.

Regression analysis of the logarithmic UO_2 data provided no strong evidence of nonlinearity or interactions between terms. Despite the modest correlation coefficient (r^2), equation (2) provides about the best possible representation of the logarithmic UO_2 data.

$$\begin{aligned} UO_2: \quad & \log(D)\{mg/m^2 \cdot day\} = \\ & 4.824 + 0.275\log_{10}[CO_3] + 0.448\log_{10}[O_2] \\ & - 0.270\log_{10}[H] - 1685/T \quad r^2=0.79 \end{aligned} \qquad (2)$$

In contrast, the spent fuel data is not adequately represented by five linear logarithmic terms as in equation (2). This is because of the complex dependence on oxygen concentration, which is illustrated in Table 2. Table 2 shows that the order of the reaction rate with respect to oxygen is approximately 0.5 for UO_2 under a l conditions tested. Spent fuel dissolution rates were almost independent of oxygen concentration at room temperature (reaction order near zero) but the reaction order was 0.73 and 0.35 for the two sets of tests performed at 75°C. Moreover, the situation may be more complex than a single interaction between oxygen concentration and temperature. Note that the two sets of tests at 75°C also had different pH and carbonate concentrations and that the reaction orders differed by a factor of two.

This complex dependence of spent fuel dissolution rate on oxygen concentration and other variables was unexpected. Therefore, some of the tests were rerun to check for repeatability. In some cases, oxygen concentrations were changed during ongoing tests while holding all other variables constant. Figure 1 is an example of marked changes in dissolution rate when the oxygen concentration was changed. Figure 2 shows almost no change in dissolution rate when the oxygen concentration was changed in a room temperature test. Because of the convincing evidence of interactions between oxygen concentration and other variables, particularly temperature, the logarithmic spent fuel data were found to be best represented by equation (3).

$$\begin{aligned} SF: \quad & \log(D)\{mg/m^2 \cdot day\} = \\ & 9.234 + 0.142\log_{10}[CO_3] - 16.73\log_{10}[O_2] \\ & + 0.140\log_{10}[H] - 2133/T + 6.81\log_{10}(T) \cdot \log_{10}[O_2] \\ & r^2=0.85 \end{aligned} \qquad (3)$$

Unfortunately equations (2) and (3) cannot be used to make comparisons between the UO_2 and spent fuel data because of their different forms. Nor can equation (3) be used to compare our spent fuel data with that of other researchers, because the complex dependence on oxygen concentration has not previously been reported. However, it is possible to make direct comparisons between our UO_2 and spent fuel data, because the specimens were tested under the same conditions, using the same test method. Comparison of test results on UO_2 and spent fuel that were obtained at LLNL and PNL, respectively, are strengthened by round-robin tests on separate portions of the same batch of UO_2 powder.[11] These tests show good agreement among the different laboratories. Finally, and this is very important, the surface areas of both the spent fuel and UO_2 powder specimens were carefully measured using the BET method. An important aspect of the BET measurement on the spent fuel specimens was that the specimens consisted of batches of separated grains.[12] This type of spent fuel specimen is crucial to the surface area measurement, because it enables the surface area that is actually contacted by water to be adequately represented by a BET measurement.

Two different averages of the spent fuel and UO_2 data were calculated (see Table 1). The first was for 20% oxygen (air) and the second was for all tests where the conditions were nominally identical. For both averages, the UO_2 rates were about three times higher than the spent fuel rates. This is a small difference for this type of test and is comparable to that observed when the two laboratories tested separate portions of the same batch of UO_2 powder.[11] Thus, there is no discernible difference between the spent fuel and the UO_2, except for the clear difference in the way the two materials responded to changes in oxygen concentration.

To allow further comparisons of the spent fuel data with the UO_2 data and with spent fuel data generated by other researchers, data at 20% oxygen were extracted from both sets and fit by equations (4) and (5). The coefficients of equations (2), ignoring the oxygen term, and (5) are similar, which supports the contention that the different variables are independent of one another. The coefficients of equations (4) and (5) are also similar, which shows that when oxygen is eliminated as a variable, spent fuel and UO_2 dissolution rates depend in a similar manner on the other variables tested. Equation (4) has the same form as one published earlier[5] and is based on essentially the same data. The coefficients are slightly different as a result of having rerun some of the tests.

SF (20% oxygen only):

$$\log(D)\{mg/m^2 \cdot day\} = 7.202 + 0.226\log_{10}[CO_3] + 0.091\log_{10}[H] - 1628/T \quad r^2=0.95. \qquad (4)$$

UO_2 (20% oxygen only):

$$\log(D)\{mg/m^2 \cdot day\} = 4.650 + 0.274\log_{10}[CO_3] - 0.187\log_{10}[H] - 1500/T \quad r^2=0.79. \qquad (5)$$

V. DISCUSSION

Aside from oxygen concentration, both spent fuel and UO_2 dissolution rates were most dependent on temperature followed by a lesser dependence on carbonate concentration. Changes in pH had the least effect on the dissolution rates of both materials, as reflected by near zero coefficients for the pH (-log[H]) terms in equations (4) and (5).

Activation energies calculated from the inverse temperature coefficients of equations (2), (4) and (5) are in the range of 6.9 to 7.7 kcal/mol. These values are in reasonably good agreement with the estimate provided by Johnson and Joling[13] of 4.5 to 6.0 kcal/mol between 25 and 150°C for spent fuel in groundwater containing approximately 0.001 mol/L carbonate.

Johnson and Shoesmith[14] reported a first-order reaction rate dependence with respect to carbonate concentrations near 0.001 mol/L and this is supported by the data reviewed by Grambow.[1] Our data suggest a lower reaction order closer to 0.25.

Johnson and Shoesmith also reported that dissolution rates were nearly independent of pH over the range 5 to 10, which agrees with our findings over the pH range 8 to 10. The data reviewed by Grambow covered the pH range of 2 to 9, and there appeared to be a trend toward increasing dissolution rates at the lower pH values. However, no clear trend existed above pH of about 6, a finding not inconsistent with our results.

Equation (2) and the data in Table 2 suggest that the reaction rate order with respect to oxygen is about 0.5 for UO_2. Grambow noted apparent general agreement among researchers that dissolution rates in acid media were linearly dependent on oxygen partial pressure. In carbonate media at high oxygen partial pressures, Grambow found that different researchers' data appeared to indicate half-order dependence on oxygen pressure. Our data support the extension of half-order dependence to oxygen partial pressures as low as 0.002 atmospheres in carbonate media.

In summary, the dissolution rates of UO_2 and spent fuel were measured as a function of four variables. All of our UO_2 data together with the spent fuel data that were obtained at atmospheric oxygen pressures appear to be in approximate agreement with published work. In sharp contrast, our finding that dissolution rates for spent fuel, but not for UO_2, are dependent upon complex interaction between oxygen concentration and temperature (plus, perhaps, pH and/or carbonate concentration) has not been reported before and requires further study for interpretation Radiolysis may account for the difference between spent fuel and UO_2 in this regard.

VI. CONCLUSIONS

The dissolution rates of spent fuel and unirradiated UO_2 were found to be about the same and to respond similarly to changes in pH, temperature, and carbonate concentration. However, the two materials responded very differently to dissolved oxygen concentration. Approximately half-order reaction rates with respect to oxygen concentration were found for UO_2 at all conditions tested. Spent fuel dissolution (reaction) rates were nearly independent of oxygen concentration at room temperature. At 75°C, reaction orders of 0.35 and 0.73 were observed for spent fuel, and there was some indication that the reaction order with respect to oxygen concentration might be dependent on pH and/or carbonate concentration as well as on temperature.

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NOMENCLATURE

a,b,c	exponents of chemical concentrations
A,B,C..	concentrations of chemical species
D	dissolution rate
E_a	Arrhenius type activation energy
k	reaction rate constant
R	gas constant
r^2	correlation coefficient
T	absolute temperature

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Table 1. Test Parameters and Results for Spent Fuel and UO_2 Dissolution Tests(a)

Run No.	Temp. (°C)	Carbonate(b) (mmol/L)	Oxygen(c) %	pH(d)	U Dissolution Rate (mg/m^2·day) SF	UO_2
1	50	2	20	9.0	6.34	
2	50	2	20	9.0	7.05	
3	50	2	20	9.0	5.07	
4	22/25	20	20	8.0/8.7	3.45	2.42
5	74/75	20	20	10.0/10.3	14.2	77.4
6	74/75	0.2	20	8.0/9.1	8.60	10.9
7	21/25	0.2	20	10.0/9.0	0.63	2.55
8	22/25	20	20	9.0/9.4	2.83	6.72
9	22/25	2	20	10.0/9.3	2.04	9.34
10	27/26	0.2	2	8.0/7.8	1.79	0.12
11	78/75	0.2	2	10.0/9.7	1.49	9.21
12	25/26	20	2	10.0/10.1	2.05	1.87
13	77/75	20	2	8.0/8.5	2.89	5.11
14	23/25	20	0.3/0.2	8.0/8.0	2.83	0.22
15	74/75	20	0.3/0.2	10.0/9.8	0.69	5.61
16	78/75	0.2	0.3/0.2	8.0/8.7	1.98	0.51
17	19/26	0.2	0.3/0.2	10.0/9.3	0.51	0.23
18	50/50	20	0.3/0.2	10.0/9.9	1.04	4.60
19	21/26	2	0.3/0.2	9.0/9.0	1.87	1.52
20	75	20	2	10.0	4.75	
21	50	2	2	8.9		12.3
22	50	2	2	8.8		7.96
23	50	2	2	8.9		10.4
24	75	0.2	20	9.5		6.48
25	75	2	20	9.6		23.3
26	75	20	20	8.5		54.0
Average	Runs 4-9				5.29	18.2
Average	Runs 4-19				3.08	8.57

(a) Numbers separated by a "/" are data for spent fuel and UO_2 respectively (SF/UO_2)

(b) Made up using appropriate amounts of Na_2CO_3 and $NaHCO_3$

(c) Percent of oxygen in sparge gas

(d) Measured at room temperature. For spent fuel, the measured values were within ±0.1 unit of the nominal values listed.

Table 2. Dependence of Dissolution Rates on Oxygen Concentration

Run Pairs	Oxygen Dependency(a) Spent Fuel	UO_2
4/14	0.047	0.53
5/15	0.73	0.62
6/16	0.35	0.66
7/17	0.051	0.52

(a) Slope of log(D) versus log[O_2] plot

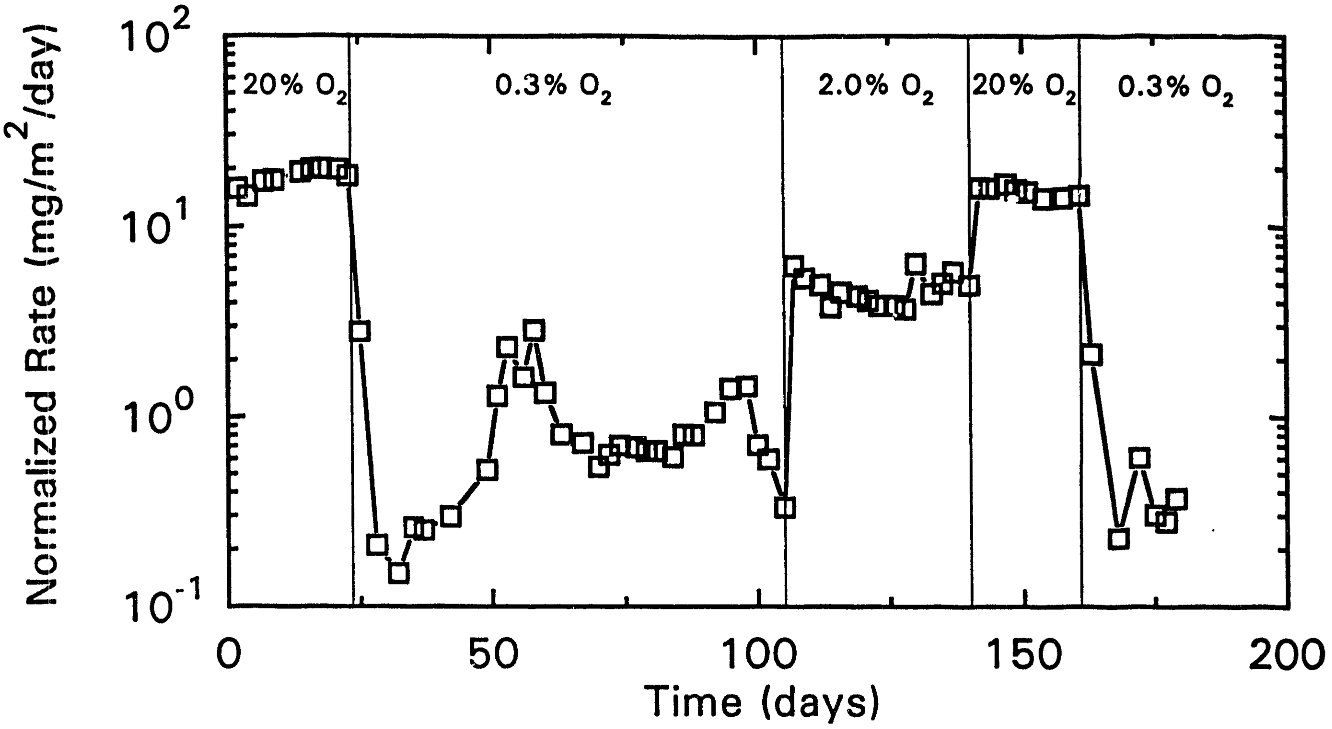

Figure 1. Spent fuel dissolution rate in 0.02 M $NaHCO_3/Na_2CO_3$, pH = 10, 75°C. Vertical lines divide regions of different O_2 overpressure (% of atmospheric).

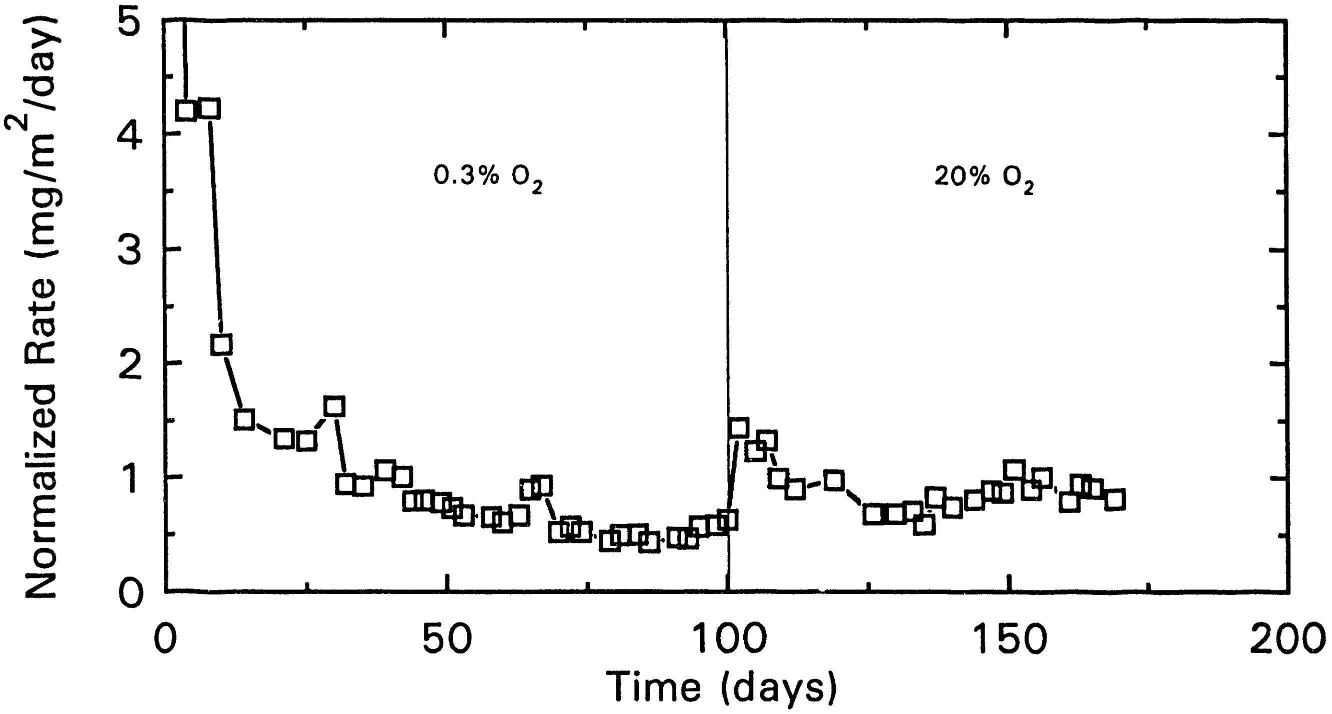

Figure 2. Spent fuel dissolution rate in 0.0002 M Na_2CO_3, pH = 10, 25°C. Vertical line divides regions of different O_2 overpressure (% of atmospheric).

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