Spent Fuel Integral Experiments

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ABSTRACT

A series of parallel spent fuel integral experiments are underway at Battelle-Columbus. These experiments are operational in the Battelle hot cell facility and are designed to provide information on effects of cladding degradation on the release of radionuclides from spent fuel waste forms and on combined effects interactions between spent fuel waste forms, the waste package, and the surrounding repository environment. To accomplish these objectives, the integral experiments have been designed the emulate characteristics of repository environments for spent fuel materials in deep-mined repositories in tuff, basalt, and granite media.

INTRODUCTION

The parallel integral experiments which are underway at Battelle-Columbus provide a means for examining interactions that may occur between components of the waste package and repository in various relational combinations. In particular, the integral experiments bring together elements of repository environments with metallic and nonmetallic components of the waste package and waste form. This approach allows for the observation of synergisms which may occur as additional degrees of freedom are introduced into the system. For example, influences of packing, corrosion products, and radiation on groundwater chemistry may induce different performance characteristics from waste package components than might be expected from experiments performed in unperturbed groundwater environments. These integral tests also provide a means to examine interactions between degraded waste package components and waste package performance; for example, interactions between radionuclide release rates and cladding degradation.

EXPERIMENTAL

The spent fuel integral experiments combine elements of waste package degradation and waste package performance in a variety of environments including packing, simulated corrosion products, basalt, and simulated groundwater. These experiments, by virtue of their principle components, are performed in a radiation field. The apparatus is assembled into two primary sections. One section, containing the spent fuel materials and other samples, is located inside a hot cell. The in-cell component is housed in an oven for operation at temperatures on the order of 90C. The remainder of the apparatus is located outside the hot cell. Since the experiment employs a once-through design, the out-of-cell portion of the apparatus provides for both the source of simulated groundwater and the collection of aqueous effluents. A matrix of experiments which are underway, and planned (those in tuff environments), is shown in Table I. In addition to the samples shown in Table I, each test section chamber, with the exception of No. 29, also contains a layer of basalt chips. As shown in the matrix of tests, the flow rates for the simulated groundwater through the test sections are one ml/day for tests 1-9 and 12-29, and ten ml/day for tests 10 and 11. Data shown in this report will reference the test numbers shown in Table I.

BURNUP ANALYSES

Part of the preoperational effort involved measuring the radionuclide inventory of representative spent fuel samples to provide reasonable estimates of the initial radionuclide content of the spent fuel waste forms used in the experiments. Part of this effort included burnup analyses of PWR and BWR spent fuel materials. These materials were excerpted from sections of spent fuel rods adjacent to locations, in the same rod, from which sections were removed for use in the integral experiments. These analyses give the ratios of the various isotopes of uranium and plutonium allowing radiochemical data to be transformed into chemical data for these elements. A precise determination of burnup also supports analytical calculations, via ORIGEN[1] or ORIGENII[2], of the composition of the material. Results of the burnup analyses of the "representative spent fuel samples" are shown in Tables II and III. Table II shows the atom percent for various uranium and plutonium isotopes as well as the $^{148}Nd/^{238}U$ and $^{239}Pu/^{238}U$ ratios. Table III shows the burnup values for the Turkey Point and Oyster Creek fuel samples taken from the rods which were used to provide the spent fuel samples for the integral experiments. Chemical analyses of the concentrations of other nuclides in the spent fuel samples are underway to provide data on the pretest inventory of radionuclides in the spent fuel specimens used in these experiments.

DATA FROM THE SPENT FUEL INTEGRAL TESTS

The spent fuel integral tests became operational on September 30, 1985. Of the 29 individual tests outlined in the test matrix shown in Table I, all are operational except number 17 as a result of difficulties encountered during sample preperation, and numbers 25-28 because the tuff material which is to be included in these test sections has not yet been received.

Since the samples were loaded in the hot cell in an air environment, the early data are not expected to be representative of the environment intended for the tests. As such, effluent from the first two months has been collected and stored for possible future analysis. Routine analyses of the effluent was initiated in December 1985. Gamma analyses for two

TABLE I

Matrix of Integral Tests

Test No.		Fractional Area Exposed (%)	Flow Rate (ml/day)
1	PWR intact fuel rod segment	0	1
2	PWR degraded fuel rod segment (1/18th inch perforations)	.5	1
3	PWR degraded fuel rod segment (1/16th inch perforations)	.5	1
4	PWR degraded fuel rod segment (1/16th inch perforations)	3	1
5	PWR degraded fuel rod segment (1/16th inch perforations)	3	1
6	PWR degraded fuel rod segment (1/16th inch perforations)	15	1
7	PWR degraded fuel rod segment (1/16th inch perforations)	15	1
8	PWR spent fuel fragments	100	1
9	PWR spent fuel fragments	100	1
10	PWR spent fuel fragments	100	10
11	PWR spent fuel fragments	100	10
12	BWR spent fuel fragments	100	1
13	BWR spent fuel fragments	100	1
14	PWR degraded fuel rod segment (1/32 inch perforations)	3	1
15	PWR degraded fuel rod segment (1/32 inch perforations)	3	1
16	BWR fuel rod section with in-service failure	-	1
17	Test cancelled--fuel fell out of sample	-	1
18	BWR fuel rod section with in-service failure	-	1
19	BWR fuel rod section with in-service failure	-	1
20	PWR sections embedded in packing (1/16-inch perforation in each of 4 spent fuel rod sections)	-	1
21	PWR cladding slit and with PWR cladding perforation embedded in packing (1/16-inch perforation and a 1/16-inch by 0.5-inch slit)	-	1
22	PWR cladding slit and with PWR cladding perforation embedded in packing (1/16-inch perforation and a 1/16-inch by 0.5-inch slit)	-	1
23	PWR fuel with cladding perforation embedded in corrosion products	-	1
24	PWR fuel with cladding slit embedded in corrosion products	-	1
25	PWR fuel fragments	-	1
26	PWR fuel fragments	-	1
27	PWR fuel fragments	-	1
28	PWR fuel fragments	-	1
29	Blank (control)	-	1
30	(not used)	-	1

TABLE II

Results of Heavy Element Analysis

Sample	Atom Percent				Nd-148 / U-238	Pu-239 / U-238
	U-234	U-235	U-236	U-238		
PWR Spent Fuel	0.015	0.600	0.346	99.039	5.648×10^{-4}	5.022×10^{-3}
PWR Spent Fuel	0.019	1.394	0.243	98.344	2.826×10^{-4}	3.981×10^{-3}

	Plutonium Isotopes - Atom Percent					Date of Analysis
	Pu-238	PU-239	P-240	PU-241	PU-242	
PWR Spent Fuel	1.617	56.393	26.437	9.488	6.065	10-23-85
PWR Spent Fuel	0.540	72.041	19.908	6.103	1.408	10-23-85

TABLE III

Calculated Burnup Values for Turkey Point and Oyster Creek Spent-Fuel Samples

Sample	F' Fission/Atom U-238	U' Atom U/Atom U-238	Pu Atom Pu/Atom U-238	F_T Atom % Burnup	Burnup MWD/MTU**
PWR Spent Fuel	3.380×10^{-2}	1.0097	8.905×10^{-3}	3.212	30.835
PWR Spent Fuel	1.691×10^{-2}	1.0168	5.526×10^{-3}	1.627	15.619

** Megawatt days per MTU.

consecutive weeks of sampling, during this period, are described in this report. These data are shown in Figures 1-3. These data show the ^{134}Cs activity in uCi per ml for the effluent collected from the sample chambers containing the tests indicated for samples collected on 12/13/85 and 12/20/85.

Figure 1 shows the data for test numbers 1-7. These tests examine influences of cladding degradation on the release rates of radionuclides. Test number one represents an intact capped fuel rod segment. Since this sample showed the highest activity of this set, and since the unit activity is dropping rapidly, we believe that these early data might be skewed by surface contamination of fine particulates which were picked up by the samples during the sectioning of the rods and final preperation of the sample specimens. Anticipating this difficulty, each fuel rod segment was superficially cleaned with a damp cloth before insertion into its test chamber. With time, the specific activities for this set, should become representative of the samples themselves.

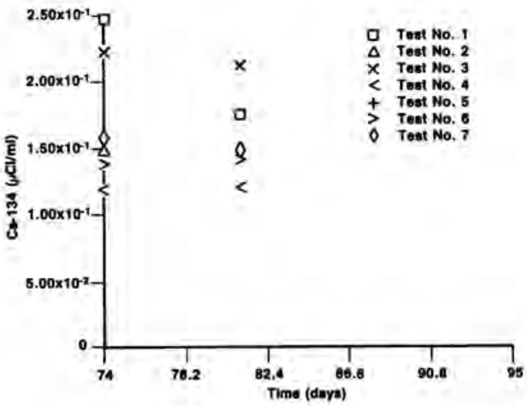

Fig 1. Specific Activity of ^{134}Cs in the Effluent from Test Numbers 1-7

Figure 2 shows the specific activities of the fluid collected from test chambers containing tests 8-13. These contain PWR and BWR fuel fragments. Since the specific activities are lower than for tests 1-7, we believe that easily dissolved material was released early on compared with the degraded fuel rod specimens. To test this, we will analyze the samples from the first two months of operation to determine the early activity releases. If sufficient activity is not found in the previous effluent from these samples, then additional explanations will be sought.

Figure 3 shows the specific activity of ^{134}Cs in the effluent from sample chambers containing test numbers 16-19. These are for sample BWR spent fuel rods containing possible in-service failures. The presence of these failures was estimated based on results of eddy current tests performed at Battelle-Columbus[3]. Activity releases were observed for test numbers 16 and 18.

SUMMARY

Spent fuel integral experiments which are underway at Battelle-Columbus have been described and some early data have been presented. These experiments examine effects of cladding degradation on the release of radionuclides from spent fuel waste forms and allows for combined effects interactions which may occur between spent fuel waste forms and other materials such as simulated corrosion products, basalt, and packing.
Elements of these experiments are applicable to spent fuel waste forms emplaced in deep-mined repositories in basalt, tuff, and granite media.

REFERENCES

1. M. J. Bell, "ORIGEN-The ORNL Isotope Generation and Depletion Code", ORNL-4628(May 1973).

2. Groff, A. G., "ORIGEN2-A Revised and Updated Version of the Oak Ridge Isotope Generation and Depletion Code", ORNL-5621, (July 1980).

3. "Long-Term Performance of Materials Used for High-Level Waste Packaging", D. Stahl and N. E. Miller (Compilers), NUREG/CR-4379, Vol. 1, First Quarterly Report, Year Four, pp4-9 to 4-28(July 1985).

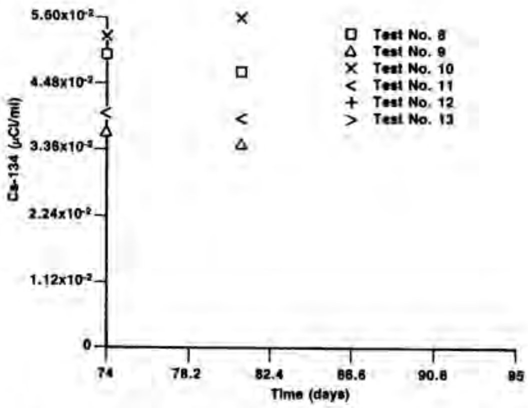

Fig. 2. Specific Activity of ^{134}Cs in the Effluent from Test Numbers 8-13

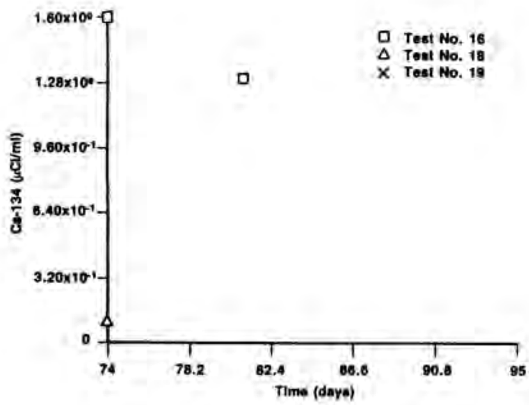

Fig. 3. Specific Activity of ^{134}Cs in the Effluent from Test Numbers 16-19