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FISSION PRODUCT RELEASE RATE FROM ALUMINUM CLAD URANIUM FUEL

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FISSION PRODUCT RELEASE RATE
FROM ALUMINUM CLAD URANIUM FUEL

INTRODUCTION

The rate of fission product inventory release to the environment as a result of exposure of uranium fuel to high temperatures should be known to more realistically assess the consequences of a credible accident. To obtain an acceptable experimental evaluation of releases it is necessary to have similar conditions to those which would exist in the reactor.

Fission product release rate experiments exploring temperatures below the uranium melting point were performed for Douglas United Nuclear by D. L. Reid of Battelle-Northwest. Data and some conclusions from the initial tests are presented in this report. The intent of distributing this information is to make known results and to record data transmitted in correspondence from Battelle-Northwest to DUN. A detailed report by Battelle-Northwest will be issued at a later date.

SUMMARY AND CONCLUSIONS

The significance of the data from these exploratory experiments is demonstrated by comparison with results from previous experiments. Although the experiments were not planned to obtain fission product release rates from molten uranium, they did indicate the need for revision of previously used values. The large clad fuels yielded significantly smaller releases than the miniature unclad uranium cylinders. This finding was consistent with tests of the N Reactor zircaloy clad fuel (performed by D. L. Reid). A much lower release rate of krypton than xenon contradicted previous assumptions that noble gas release rates were equal. This was also observed for the zircaloy-2 clad N Reactor Mark I fuel tests. Cesium and iodine releases were also at a much lower rate than those predicted from the miniature specimen tests.

During the experimentation significant exothermic reactions of aluminum with uranium were observed at the β - γ uranium phase change and a second reaction at ~ 1080 C creating localized temperatures in the range of 1500 C, predominately near the end caps.

Previous experiments with miniature unclad specimens showed that irradiation differences of the magnitude used in these full-size element tests would not make a significant difference in the fission product release rates.⁷ The use of an air atmosphere in one test did not significantly alter the release obtained under a steam atmosphere - probably because of the small percentage oxidation of the uranium.

DISCUSSION

Data are needed for use in predicting the fission product release rate for postulated coolant loss accident cases for the low pressure reactors. Laboratory experiments to determine the fission product release rate were run using full-size, aluminum-clad fuel elements heated to simulate the computed temperature transient shown in figure 1. Due to limitations in the facility and sampler shielding, the experiments were restricted to thermal temperatures below the uranium melting point to minimize personnel exposure problems.

Figure 2 describes the general setup of the experimental apparatus. A single fuel element was heated in a steam atmosphere by an induction coil surrounding a horizontal quartz furnace as shown in Figure 3. Removal of the steam after passage through the furnace was accomplished by a condenser and a cold trap. To carry the fission products through the system, air was injected into the condensate flask. All the fission products except the noble gases were removed by a May Pack sampler and the condensate. Noble gases were collected principally in -70 C charcoal traps.

Table 1 is a compilation of data from all experiments run to date. The data should be considered as indicating the range of release to be expected for fuel which does not melt (tests 1, 2 and 3) or partially melts (tests 4, 5 and 6). The number of tests has been limited by the cost and exposure involved in using the available facility and by similar tests using the N Reactor fuel. The variation in release data between tests was due to differences in test conditions. These were the first tests of this kind to be run and the limited shielding available for the samplers dictated that caution be used to prevent melting and the potential large fission product release to the samplers. In addition, on discovering that a rapid reaction was initiated at about 1080 C, further exploratory test variations were made to determine the nature of the reaction and confirm the initiation temperature.

"Burst" is defined as the release from 650 C to goal temperatures³, except for test 2. For the second test, "burst" is defined to 670 C, the second sampling period from 670 to 810 and the third period from 810 to termination of the test after 20 minutes at 1010 C. During the constant temperature period two at temperature samples were taken over variable times. The time vs. temperature plot of Figure 1 indicates the approximate data collection pattern. The percent inventory value is computed by dividing the amount of fission product isotope released during the period indicated by the computed inventory of that isotope. Because measurement could not be made, the percentage melt values are only estimates based on post heating examination.

Total release values recorded in Table 1 are much lower than those of the miniature unclad samples (Table 2) for all the fission products measured. Since the experimental procedures were similar it is evident that mass and cladding are factors affecting the release for the no melt and partial melt cases. Additional planned experiments with different fuels will provide

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comparable data both below and above the melting point of uranium to more accurately assess the effect of mass and cladding on release rates as well as fractional releases. It was assumed in the past that the release of fission products is the same for all noble gases. The data show a significantly higher release of xenon than krypton for the aluminum clad fuel tests. All tests indicate these contrasting release characteristics for the two noble gases, but for the completely molten uranium case their release should be equal. Similar observations were made for the N-fuel tests.

The data show that iodine is released at a low rate which is a relatively similar rate at all test conditions. This indicates that the iodine release rate is primarily related to the uranium oxidation rate for these test conditions. Little oxidation appeared to occur in the tests. An air atmosphere test was included in the series as an exploratory step. This did not show a substantial variation from the steam results, which is probably due to the small amount of uranium oxidation.

Currently planned tests will measure the hydrogen released when aluminum-clad uranium is heated in both the unirradiated and the irradiated states; this is a method of determining the extent of uranium oxidation by the steam atmosphere. Full melt tests will be performed later this year after modification of a hot cell in the 324 building is completed. Tests will also be run to determine the effect of aluminum process tubes on the uranium-aluminum reaction heating. Since nickel-clad fuel may be used extensively in the future a few of these will also be tested.

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FIGURE 1. TEST TEMPERATURE CYCLES

FIGURE 2. TEST EQUIPMENT ARRANGEMENT

FIGURE 3. INDUCTION FURNACE ARRANGEMENT

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TABLE 1

FISSION PRODUCT RELEASE EXPERIMENTS

Al-CLAD URANIUM FUEL, Al-Si BONDED

Test No. Isotope Release Sample	<u>1</u> %Inv Time*	<u>2</u> %Inv Time*	<u>3</u> %Inv Time*	<u>4</u> %Inv Time*	<u>5</u> %Inv Time*	<u>6</u> %Inv Time*
<u>Xenon</u>						
Burst	0.1 11	0.08 5	0.5 12	0.4 5	0.3 5	1.0 5
At Temp. 1st	0.3 } 10	0.012 } 23	0.3 } 15	18.0 } 5	11.2 } 11	24.8 } 8
2nd	3.20	1.7	0.6	2.9	5.4	1.2
Cool down	0.0	2.2	7.6	3.8	4.8	0.9
Total	3.6	4.0	9.0	25.1	21.7	27.9
<u>Krypton</u>						
Burst	0.02	0.0001	0.2	0.15	0.09	0.3
At Temp. 1st	0.14	0.004	0.08	8.3	5.9	14.5
2nd	0.9	.375	0.07	1.4	2.9	6.4
Cool down	.15	0.25	0.2	0.15	0.5	0.2
Total	1.21	0.629	0.55	10.0	9.4	21.4
<u>Iodine</u>						
Burst	0.03	0.204	0.07	0.01	0.01	0.11
At Temp. 1st	0.17	0.002	0.1	.20	0.23	0.04
2nd	0.3	0.335	0.13	.09	0.26	0.44
Cool down	0.0	0.0	0.0	0.0	0.0	0.0
Total	0.5	0.60	0.30	0.3	0.5	0.59
<u>Cesium</u>						
Burst	0.00308	0.0009	0.008	0.001	0.01	0.11
At Temp. 1st	0.008	0.0001	0.007	0.081	0.14	0.97
2nd	0.012	0.022	0.005	0.08	0.10	0.07
Cool down			0.0	0.0	0.0	0.0
Total	0.02	0.023	0.020	0.1	0.25	1.15
<u>Conditions</u>						
Atmosphere	Steam	Steam	Steam	Steam	Steam	Air
Melting	0	0	0	30	10	50
NVT x 10 ⁻²⁰	2.2	2.2	3.0	3.0	3.0	3.0
Gcal Temp.	1015	1015	1015	1100	1100	1100

* Times apply to all isotopes.

TABLE 2

MINIATURE UNCLAD FUEL EXPERIMENTS, FISSION PRODUCT RELEASE³

T = 1000 C for 45 min.

Irradiated to 1.2×10^{19} nvt

<u>Isotope</u>	<u>% of Inventory</u>
¹³¹ Xe	99
⁸⁵ Kr	--
¹³¹ I	84
¹³⁷ Cs	55
¹²⁹ Te	44

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