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UC-40 Radiation Effects on Materials

THREE-MINUTE IRRADIATION AT VARIABLE POWER DENSITY OF PROTOTYPE CETR FUEL PELLETS

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Printed in USA. Price $0.50 . Available from the

Office of Technical Services
Department of Commerce
Washington 25, D.C.

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ORNL-2999

Contract No. W-7405-eng-26

CHEMICAL TECHNOLOGY DIVISION

CHEMICAL DEVELOPMENT SECTION B

THREE-MINUTE IRRADIATION AT VARIABLE POWER DENSITY OF PROTOTYPE CETR FUEL PELLETS

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SEP 28 1960

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ABSTRACT

Prototype CETR Fuel pellets (95% ThO_2-UO_2, 93% of theoretical density) were irradiated 3 min in the ORR at power densities ranging from 16 to 160 Mw/metric ton Th. No fragmentation or cracking of the pellets occurred. Variations in the irradiation conditions had no effect on the rate of dissolution of the pellets in boiling 13 M HNO_3—0.04 M NaF—0.1 M $Al(NO_3)_3$.

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1.0 INTRODUCTION

Both the Darex-Thorex[1,2] and Sulfex-Thorex[1,3] processes for the dissolution of Consolidated Edison (CETR) fuel involve removal of the stainless steel jacket prior to dissolution of the 95% ThO_2-UO_2 pellets in 13 M HNO_3—0.04 M NaF—0.1 M $Al(NO_3)_3$. Fragmentation of the fuel pellets during irradiation not only could lead to higher decladding losses but also could result in the formation of slurries of fine core material, which would constitute a criticality hazard.[4] Irradiation of ThO_2-UO_2 pellets by other workers[5] has led to fragmentation in some cases but not in others.

The objective of the current experiments was to determine the power density required to crack 0.26-in.-dia 0.5-in.-long 95% ThO_2-UO_2 pellets (93% of theoretical density) which are similar to those which will be used in the first CETR core. Since the thermal gradient within a fuel pellet is established almost immediately after its insertion into a reactor, the specimens were irradiated only 3 min in the ORR so that bulky shielding could be avoided in subsequent operations. The power densities ranged from 16 to 160 Mw/tonne Th. Of primary interest was the power density (or temperature gradient) at which cracking of the pellets occurred, the amount and size distribution of fines produced, and the effect of power density on the rate of dissolution of the fuel pellets in 13 M HNO_3—0.04 M NaF—0.1 M $Al(NO_3)_3$.

The authors are pleased to acknowledge the assistance of J. P. Nichols of the Chemical Technology Division who designed the irradiation capsules. Chemical, x-ray, spectrographic, and particle size analyses were made by the groups of G. R. Wilson, W. R. Laing, R. L. Sherman, C. Feldman and G. W. Leddicotte of the ORNL Analytical Chemistry Division.

2.0 PROCEDURE

Each of the twelve tubular aluminum irradiation capsules (or rabbits) which were fabricated contained three 95% ThO_2-UO_2 pellets (~12 g).[6] In addition to the ThO_2-UO_2 pellets, each rabbit contained two high-density MgO insulating pellets (~1 g) and a piece of cobalt wire (~1 g) for flux determination. Details of the rabbits are given in Drawing D-33959.

The capsules were irradiated in pairs for 3 min in the ORR at 1, 2, 4, 6, 8, and 10 Mw. The aluminum cladding was then removed chemically and the MgO pellets and cobalt wire removed manually to allow inspection of the fuel pellets. All post-irradiation experiments were conducted in the shielded cell in Room 19, Building 4505. The aluminum was dissolved in boiling 2 M NaOH—1.78 M $NaNO_3$. Solids produced in this step, resulting from the iron impurity in the aluminum, were washed three times with 1 M NaOH before their dissolution in boiling 0.5 to 1 M HCl. The HCl was then removed in several water washes. Extreme care was taken to leave all solids in the reaction vessel. Consequently, all the fuel material was ultimately obtained for weighing, particle size measurement, and dissolution studies.

3.0 RESULTS

3.1 Neutron and Heat Flux Data

Fluxes obtained from the Co wire monitors in the rabbits were slightly lower than those anticipated from J. W. Ullmann's calculations[6] (Table 1). The specific powers produced did span the range of 16 to 160 Mw/tonne Th expected in the CETR, and the peak central pellet temperature was in the neighborhood of 2500°C, still far below the melting point of the pellets.

3.2 Particle Size Data

During the 3-min irradiation period no fracturing of the pellets occurred. In all cases a small amount of fine, powdered material was found which could have been caused by chipping and spalling while handling the capsules.

Table 1. Apparent Thermal Neutron Fluxes in 3-min Irradiation Tests

ORR Power, Mw	Flux x 10^{-13} at Surface of Pellets, $n/cm^2/sec$		Approximate Specific Power, Mw/tonne Th
	Calculated[6]	Measured	
1	0.94	0.808	16
1	0.94	0.856	16
2	1.88	1.76	32
2	1.88	1.73	32
4	3.76	3.62	63
4	3.76	3.59	63
6	5.64	5.48	94
6	5.64	5.72	94
8	7.52	7.91	125
8	7.52	7.93	125
10	9.40	8.88	157
10	9.40	9.08	157

The amount of fines was less than 0.02 wt % except in one capsule which was dropped during handling prior to the chemical tests (Table 2). The fines from this capsule represented 0.4% of the original pellets. Except in the case of the dropped capsule, greater than 80% of the fines were less than 10 microns in size (Table 2). Since no gross fracturing of the pellets occurred, the particle size data are not significant.

Table 2. Amount and Size of Fuel Fines Formed in 3-min Irradiations of 95% ThO_2-UO_2 Pellets

Capsule No.	ORR Power, Mw	Fines Produced, %	Amt. of Fines >10 μ, %
6	1	0.006	-
1	1	0.005	-
3	2	0.009	-
7	2	0.010	-
2	4	0.010	-
4	4	0.011	-
10*	6	0.37	87
12	6	0.015	18
8	8	0.019	1
11	8	0.002	10
5	10	0.014	10
9	10	0.004	14

*This capsule dropped during handling.

3.3 Chemical Data

The fuel pellets were supplied by Nuclear Metals and Equipment Company and were to contain 4.4% UO_2 (93% enriched uranium). Their densities were to be nominally 93% of theoretical. By analysis at ORNL, the following composition was determined: O/U = 2.05 or less; % UO_2 = 4.67; % Al = 0.1; density = 93-94% of theoretical.

Dissolution tests to determine whether irradiation conditions affected the dissolution rate were conducted as follows: The pellets were first digested in glass vessels for 5 hr in a 200% stoichiometric excess of boiling 13 M HNO_3--0.04 M NaF--0.1 M $Al(NO_3)_3$. Following this treatment were three more 5-hr digestions, each with 1/2 volume of fresh reagent. Then the residue was weighed and analyzed.

In the first digestion an average of 88.5% of all irradiated pellets dissolved irrespective of the power density which had been established (Table 3). Only 80% of a batch of the unirradiated pellets dissolved in a similar test. It is doubtful that irradiation caused this difference since the amount dissolved in 2 digestions (~99.9%) was essentially the same in all cases. A white residue was always present after four digestions. This residue was shown by x-ray and chemical analyses to be mainly Al_2O_3 and 6 $Al_2O_3 \cdot CaO$. The weight of the residue was about 0.2% of the original pellets (Table 4). Analyses of randomly selected residues revealed that a maximum of 0.0002% of the uranium and thorium was present in the solids.

Table 3. Dissolution of 95% ThO_2-UO_2 Pellets Which Were Irradiated 3 min in the ORR

Dissolvent: boiling 13 M HNO_3—0.04 M NaF—0.1 M $Al(NO_3)_3$
First Digestion: 200% stoichiometric excess of reagent; 5 hr
Subsequent Digestions: 1/2 volume of fresh reagent; 5 hr

Capsule No.	ORR Power, Mw	Amount Dissolved, % 1st Digestion U	1st Digestion Th	2nd Digestion U	2nd Digestion Th	Material Balance, %
0	0	80.1	79.3	20.0	20.4	98.8
3	2	87.2	85.7	12.6	14.1	95.5
7	2	89.7	88.7	10.3	11.3	98.3
2	4	85.8	84.8	14.1	15.0	97.3
4	4	89.9	90.0	10.0	9.9	100.1
10	6	92.7	92.3	7.3	7.6	98.7
5	10	86.8	88.9	13.1	11.0	97.3

Table 4. Residues Obtained After Four Digestions with Boiling 13 M HNO_3—0.04 M NaF—0.1 M $Al(NO_3)_3$

Capsule No.	Amount of Residue,[a] %	Losses to Residue, % U	Losses to Residue, % Th
0	0.43	-	-
3	0.17	-	-
7	0.13	0.00019	0.00015
2	0.17	-	-
4	0.09	0.00021	0.000098
10	0.15	-	-
5	0.17	0.0002	0.0001

[a]Residue was mainly Al_2O_3 and 6 $Al_2O_3 \cdot CaO$.

4.0 REFERENCES

1. L. M. Ferris and A. H. Kibbey, "Sulfex-Thorex and Darex-Thorex Processes for the Dissolution of Consolidated Edison Power Reactor Fuel: Laboratory Development," ORNL-2934, in press.
2. L. M. Ferris, "Decladding of Consolidated Edison Power Reactor Fuel by Sulfex and Darex Processes: Cyclic Dissolution Experiments," ORNL-2822 (Jan. 13, 1960).
3. L. M. Ferris and A. H. Kibbey, "Laboratory Development of the Sulfex Process for the Dissolution of Consolidated Edison Power Reactor Fuel," ORNL-2714 (Oct. 16, 1959).

4. A. R. Irvine, "Special Problems in Chemical Processing at ORNL," in Proceedings of the AEC Symposium for Chemical Processing of Irradiated Fuels from Power, Test and Research Reactors," TID-7583 (January 1960) p. 420.

5. J. C. Deddens, Babcock and Wilcox Company, Seminar presented before the ORNL Chemical Technology Division (Jan. 26, 1960).

6. J. W. Ullmann, "Computations for "Neutcracker" Experiment in ORR Hydraulic Facility," ORNL-CF-59-11-58 (Nov. 17, 1959).

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TID-4500 (15th ed.)

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