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WAPD-TM-159

AEC RESEARCH AND
DEVELOPMENT REPORT

RADIOCHEMISTRY OF THE PWR FUEL MATERIAL CYCLING TESTS (WAPD-29-1 and -2) IN THE WAPD-29 VH-3 LOOP OF THE MATERIALS TESTING REACTOR

NOVEMBER 1959

CONTRACT AT-11-1-GEN-14

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November 1959

Contract AT-11-1-GEN-14

Price \$.75

**Available from the Office of Technical Services,
Department of Commerce,
Washington 25, D. C.**

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This report presents the results of in-pile irradiation tests of PWR fuel materials with particular regard to the chemical and radiochemical aspects. These tests were conducted to study the difference of fission product release from defected fuel specimens containing UO_2 fuel materials of two different specific densities; also, one test included a comparatively high oxygen concentration in the loop water to study the effect of an accidental oxygen addition on the rate of fission product release. No firm deduction could be drawn as to the effect of different fuel densities on the emanation of fission products from defected specimens, and the high oxygen concentration had practically no effect on the rate of fission product release.

RADIOCHEMISTRY OF THE PWR FUEL MATERIAL CYCLING TESTS (WAPD-29-1 AND -2) IN THE WAPD-29 VH-3 LOOP OF THE MATERIALS TESTING REACTOR

R. Ehrenreich

I. INTRODUCTION

This is the fifth report (Refs 1, 2, 3, and 4) in a series which presents the results of in-pile irradiation tests of PWR fuel materials with particular regard to the chemical and radiochemical aspects. The purpose of this phase of testing was to study the difference of fission product release from defected fuel specimens containing UO_2 fuel materials of two different specific densities. These tests were run in the WAPD-29 VH-3 Loop of the Materials Testing Reactor (MTR), Idaho Falls, Idaho. They started on December 1, 1956, and were concluded on March 10, 1957. A general summary of all defect tests of PWR fuel elements up to September 1957 is given in Ref 5.

II. DESCRIPTION OF TEST

A. Description of Test Facility

The test loop is a conventional high temperature-pressure water facility previously utilized in a fuel materials experiment. A unique feature of this facility was an automatic mechanism for the cycling of fuel specimens in and out of the high neutron flux region of the reactor. All portions of the in-pile pressure tube (1/2-in., schedule 40 pipe) in contact with the loop water were made of type 304 stainless steel. The pressure tube is a once-through type, with primary water entering from the sub-pile room flowing upward over the specimen train and returning through a 1-in., schedule 80 pipe to the sub-pile room to complete the water circuit. A simplified schematic of the main loop and purification side stream auxiliary loop is shown in Fig. 1.

The cycling mechanism, which provided a maximum specimen-train travel of 21 in., was actuated by a high-pressure pump. A metered quantity of water was pumped into one side of a cylinder containing a piston to which the specimens were attached. This moved the piston in one direction; a pressure relief valve allowed the piston to return to its original position. An automatic timer which controlled solenoids was used to actuate the pump and relief valve alternately and thus determine the intervals of high and low neutron flux irradiation. A magnetic rod attached to the piston was placed

Fig. 1 Simplified Schematic of Main Loop and Purification Loop

within an inductance coil and used as a position indicator. A detailed description of the cycling mechanism can be found in Ref 6.

B. Description of Test Specimens

The WAPD-29-1 in-pile test section contained two PWR reference UO_2 fuel rod specimens. One specimen was predefected with a 5-mil (0.005 in.) hole approximately 4.35 in. from the top of the fuel compartment. The other specimen was undefected. The fuel material was MCW*, natural UO_2 powder which was compacted and sintered at the Bettis Laboratory. The specimens were fabricated from UO_2 pellets which averaged 93% of theoretical UO_2 density.

The WAPD-29-2 in-pile test section was identical in design and structure with that just described, except that the specimens were made from fuel pellets which averaged 98% of theoretical maximum UO_2 density.

Table I lists detailed measurements of the WAPD-29-1 and -2 test specimens and includes exact fuel densities and fuel-cladding clearances. A schematic diagram of the test specimen assembly is shown in Fig. 2.

C. Test Operations and Loop Conditions

The irradiation of the low density fuel specimens in the high flux region of the reactor began with reactor startup at 0510 hr on December 1, 1956. The loop water was maintained at a pressure of 2200 psig and a bulk water temperature of 550°F. The chemistry conditions of the coolant were as follows:

- 1) Oxygen concentration less than 0.1 ppm (except for the scheduled high oxygen concentration phase of the test).

* Mallinckrodt Chemical Works, Mallinckrodt Chemical Company, St. Louis, Mo.

TABLE I
DESCRIPTION OF TEST SPECIMENS - WAPD-29-1 AND -2

Test	Specimen*	Defect (in.)	UO ₂ Density** Range	Fuel Clearance Diam (in.)	Ave Pellet Diam (in.)	Fuel Weight (g)	Fuel Length (in.)
WAPD-29-1	LD-D	0.005	93.07-93.89	0.003-0.004	0.3566	151.567	9.089
	LD-U	None	93.16-94.16	0.003-0.004	0.3566	151.579	9.083
WAPD-29-2	HD-D	0.005	97.17-97.63	0.003-0.005	0.3561	157.434	9.079
	HD-U	None	97.04-98.02	0.003-0.005	0.3561	157.780	9.098

* LD-D = Low density, defected specimen
LD-U = Low density, undefected specimen
HD-D = High density, defected specimen
HD-U = High density, undefected specimen

** Percent theoretical UO₂ density.

Fig. 2 Schematic Diagram of Test Specimen Assembly

- 2) Hydrogen concentration maintained at 25-35 cc by gas additions (except for the scheduled high oxygen concentration phase of the test).
- 3) Loop water pH 9.5-10.5; achieved by the addition of LiOH to the loop water and maintained by a LiOH form, ion-exchange side-stream purification resin.
- 4) Conductivity; corresponding to pH.

Operations during the first two reactor cycles with the specimens in the high flux position for 27.1 full power days were normal, except that within the first five days of test operation the reactor had approximately 20 scrams which is unusually high for the MTR facility. General loop and specimen parameters are given in Table II. During the third reactor cycle, the specimens were moved into the low flux region of the facility. When a new purification resin was valved on stream at the beginning of the third irradiation cycle, improper installation prevented proper functioning for approximately 8 of the 17 days of operation (13.4 full power days of reactor operation); consequently, there was no loop water demineralization for this period.

Irradiation of the high density fuel specimens in the high flux region of the reactor commenced at 1045 on February 1, 1957. With the exception of an accidental loop drain on February 2 which probably caused boiling and a temperature excursion in the in-pile pressure tube, but resulted in no

TABLE II
GENERAL LOOP AND SPECIMEN PARAMETERS

	WAPD-29-1 Low Density UO ₂		WAPD-29-2 High Density UO ₂	
	High Flux	Low Flux	High Flux	Low Flux
Test phase started (hr-date)	0510-12/1/56	0739-1/11/57	1045-2/1/57 1745-3/4/57*	1435-2/23/57
Test phase terminated (hr-date)	0730-1/6/57	0215-1/28/57	0400-2/18/57 2315-3/10/57	1745-3/4/57
Power output defected specimen (Kw)	4.97	0.45	6.15	0.33
Burnup, MWD/T _{UO₂} of defected specimen	752	25	515	236
Ave power produced by test section (Kw)	9.26	2.3	11.45	2.95
Max. heat flux (Btu/hr-ft ²)	2.3×10^5	2.1×10^4	2.9×10^5	2.7×10^4
Megawatt days of MTR Reactor	1083	535	846	338
Fissioning rate of defect specimens (fissions/sec)	1.78×10^{14}	1.61×10^{13}	2.20×10^{14}	1.18×10^{13}
Equivalent time at full power (days)	27.1	13.4	21.2	8.4
pH	9.5-10.5	9.5-10.5	9.5-10.5	9.5-10.5
Total time of test (hr)	665.28	230.56	470.56	271.39
Ave bulk water temp (°F)	550	546	550	546
Ave water temp test section inlet, (°F)	545	545	545	545
Ave water temp test section outlet, (°F)	557	548	562	549
Total volume, V _o , of system (g)	3.82×10^4	3.82×10^4	3.82×10^4	3.82×10^4
v _p , Ave purification stream flowrate (gpm)	0.06	0.06	0.06	0.06
v _o , Ave main loop flowrate (gpm)	5.0	5.0	5.0	5.0
$\beta = v_p/V_o$, purification constant ave (sec ⁻¹)	9.908×10^{-5}	9.908×10^{-5}	9.908×10^{-5}	9.908×10^{-5}
ϕ = Ave perturbed neutron flux Calculated from power produced (n/cm ² -sec)	1.18×10^{14}	1.07×10^{13}	1.41×10^{14}	7.55×10^{12}

* Specimens returned to high flux position until end of test.

apparent damage to either the specimen train or the facility, the test progress continued satisfactorily to the end of the test's first reactor cycle (14.9 full power days). The low flux irradiation cycle began at 1435 on February 23, 1957 and continued to run at reference water conditions and low neutron flux until 1745 on March 4, 1957 when the specimens were returned to the high flux position of the reactor for an additional 6.3 full power days of operation. At 0200 on March 7, 1957, an average oxygen concentration of 12 cc/kg was introduced to the loop water. The concentration of oxygen was

maintained in the loop for 32 hr until 0930 March 8, 1957 when the reference chemistry conditions were restored. The comparatively high oxygen concentration was maintained in the loop to study the effect of an accidental oxygen addition on the rate of fission product release. The fuel specimens remained in the high flux position of the reactor and the loop water was maintained at reference conditions until the end of the test at 2315 on March 10, 1957.

III. SUMMARY OF TEST RESULTS

Although the average rate of fission product evolution, in general, was higher in WAPD-29-1 than in WAPD-29-2, the test values are greatly variable. Loop water activities of Kr^{88} and Cs^{138} were lower than expected when the specimens were in the low flux irradiation phases of the test.

The presence of a high oxygen concentration in the loop water during the test did not appear harmful to the defected fuel specimen or alter the rate of evolution of fission products.

IV. CHEMICAL AND RADIOCHEMICAL OBSERVATIONS

A. Loop Water Chemistry

Operational records reveal no deviations from the specified loop water chemistry conditions previously listed. The demineralizer resins were off stream for brief periods of time because of operational difficulties, as previously noted under "Test Operations and Loop Conditions." The data indicated that the pH averaged 10.2 when the oxygen concentration was nil. However, when the oxygen in the loop water was increased to an average concentration of 17.4 ppm (12.4 cc/kg) the average pH of the loop water was 9.4. Table A-I of the Appendix presents loop water chemistry data for the period just prior to and during the accidental oxygen addition phase of the test.

B. Radiochemistry Observations

Steady-state specific activities of Kr^{88} , I^{133} , Cs^{138} , Ba^{139} , I^{131} , and other nuclides were determined in loop water at frequent intervals during the progress of the test (Tables III and IV).

The efficiency of the LiOH resin ion exchange decontamination of loop water was investigated. Samples taken before and after the ion exchange column were analyzed for I^{131} , Cs^{137} , Ba^{139} , I^{133} , and Np^{239} (Table V).

Three purification resins from WAPD-29-1 and four resins from WAPD-29-2 were analyzed for I^{131} , Cs^{137} , Sr^{89} , Sr^{90} , Ba^{140} , Zr^{95} , and Ce^{144} (Table VI).

V. POST-IRRADIATION OBSERVATIONS

Defected specimens of the WAPD-29-1 and WAPD-29-2 tests were examined macroscopically and microscopically, and did not show any obvious abnormalities of the cladding or fuel material which could be attributed unequivocally to an irradiation effect. The usual pellet fractures were observed. Therefore, these tests could be placed in the category of so called "normal" tests defined in Ref 3 as tests in which the UO_2 pellets exhibit essentially no effect of irradiation such as melting, coring, cladding failures, etc.

VI. EVALUATION OF DATA

The escape rate coefficient ν was calculated from isotopic activity both on resins and from steady-state loop water samples. The escape rate coefficient ν is defined as the proportionality constant which relates the total amount of an isotope in the fuel to its rate of escape to the external system. This is expressed by the differential equation

$$R = \nu a(N^a)_f$$

TABLE III

SUMMARY OF STEADY-STATE ACTIVITIES, WAPD-29-1

Loop Water Activities (dis/min-ml)

Date	Time	Kr ⁸⁵	Cs ¹³⁸	Cs ¹³⁹	I ¹³¹	I ¹³³	Ba ¹³⁹	Ba ¹⁴⁰	Cs ¹³⁷	Np ²³⁹	Xe ¹³⁸
12-1-56	0510	--	--	Reactor startup, low density specimens in high flux position)					--	--	--
12-3-56	1005	--	8.38x10 ⁶	--	--	--	6.6x10 ⁴	--	--	--	--
12-4-56	0840	--	7.02x10 ⁶	--	--	--	6.91x10 ⁴	--	--	--	--
12-5-56	1000	3.54x10 ⁷	8.10x10 ⁶	--	9.64x10 ⁴	8.30x10 ⁵	1.21x10 ⁵	3.69x10 ¹	6.66x10 ¹	--	--
12-6-56	0825	--	7.23x10 ⁶	--	9.26x10 ⁴	1.15x10 ⁶	1.32x10 ⁵	3.18x10 ¹	2.26x10 ¹	--	2.79x10 ⁶
12-7-56	1124	3.13x10 ⁷	--	--	--	--	--	--	--	--	5.24x10 ⁶
	1255	--	9.42x10 ⁶	--	8.80x10 ⁴	8.4x10 ⁵	1.32x10 ⁵	--	--	--	--
12-10-56	0949	3.13x10 ⁷	7.68x10 ⁶	--	4.33x10 ⁴	5.45x10 ⁵	9.43x10 ⁴	--	--	--	--
12-13-56	1359	--	7.65x10 ⁶	--	3.63x10 ⁴	3.25x10 ⁵	9.16x10 ⁴	--	2.53x10 ¹	--	--
12-14-56	1021	3.85x10 ⁷	--	--	--	--	--	--	--	--	--
	1421	--	6.91x10 ⁶	--	2.25x10 ⁴	2.3x10 ⁵	7.93x10 ⁴	--	--	3.21x10 ³	--
12-26-56	--	--	--	2.69x10 ⁵	--	--	--	--	--	--	--
12-31-56	--	--	--	3.84x10 ⁵	--	--	3.12x10 ⁴	--	--	--	--
1-2-57	0851	1.52x10 ⁷	--	--	--	--	--	--	--	--	--
1-3-57	1241	--	4.71x10 ⁶	--	--	--	--	--	--	--	--
1-11-57	0739	--	--	--	--	(Specimens in low flux position)			--	--	--
1-12-57	0953	--	2.57x10 ⁴	5.57x10 ³	--	--	--	--	--	--	--
1-14-57	0937	--	3.79x10 ⁴	9.02x10 ³	--	--	--	--	--	--	--
1-16-57	1249	--	3.31x10 ⁴	7.98x10 ³	--	--	--	--	--	--	--
1-17-57	1407	--	2.77x10 ⁴	--	--	--	--	--	--	--	--

TABLE III (Cont)

Loop Water Activities (dis/min-ml)

Date	Time	Kr ⁸⁸	Cs ¹³⁸	Cs ¹³⁹	I ¹³¹	I ¹³³	Ba ¹³⁹	Ba ¹⁴⁰	Cs ¹³⁷	Np ²³⁹	Xe ¹³⁸
1-18-57	1430	--	2.46x10 ⁴	--	--	--	--	--	--	--	--
1-21-57	1041	1.41x10 ⁵	2.78x10 ⁴	7.02x10 ³	1.13x10 ⁴	2.90x10 ⁴	8.57x10 ²	--	--	--	--
1-22-57	0925	1.79x10 ⁵	2.71x10 ⁴	8.93x10 ³	--	3.95x10 ⁴	8.90x10 ²	--	--	--	--
1-23-57	0825	1.91x10 ⁵	3.07x10 ⁴	9.53x10 ³	1.27x10 ⁴	--	--	--	--	--	--
1-24-57	0938	1.52x10 ⁵	3.95x10 ⁴	--	1.35x10 ⁴	--	9.01x10 ²	--	--	--	--
1-28-59	0215	--	--	(Reactor shutdown, end of WAPD-29-1 irradiation)				--	--	--	--

TABLE IV
SUMMARY OF STEADY-STATE ACTIVITIES, WAPD-29-2

Date	Time	Kr ⁸⁸	Cs ¹³⁸	Cs ¹³⁹	I ¹³¹	Ba ¹³⁹	Comments
2-1-57	1045	--	--	--	--	--	Reactor startup high density specimens in high flux position
	1732	--	8.59x10 ⁶	--	--	--	
2-2-57							Accidental loop drain and temperature excursion
2-4-57	1014		2.71x10 ⁶	3.60x10 ⁵	--	--	
2-6-57	0948	6.92x10 ⁶	2.79x10 ⁶	--	--	--	
2-7-57	1008	--	2.80x10 ⁶	3.91x10 ⁵	--	--	
2-8-57	0946	--	3.82x10 ⁶	5.62x10 ⁵	--	7.00x10 ⁴	
2-12-57	1456	--	2.17x10 ⁶	--	2.13x10 ⁴	--	
2-13-57	1121	9.56x10 ⁶	--	--	--	--	
	1247	--	--	--	1.70x10 ⁴	--	
2-23-57	1435	--	--	--	--	--	Specimens moved to low flux position
2-25-57	0938	6.81x10 ⁴	3.85x10 ⁴	--	--	--	
2-26-57	0903	7.58x10 ⁴	3.70x10 ⁴	--	--	--	
3-1-57	0911	--	1.08x10 ⁵	--	--	--	
3-4-57		--	--	--	--	--	Specimens returned to high flux position
3-5-57	1213	2.56x10 ⁶	1.37x10 ⁶	--	--	--	
3-6-57	1028	2.69x10 ⁶	1.05x10 ⁶	--	--	--	Oxygen (~12cc/kg) introduced to loop
	1313	--	1.22x10 ⁶	--	--	--	
	1318	3.54x10 ⁶	--	--	--	--	
3-7-57	1539	3.11x10 ⁶	1.20x10 ⁶	--	--	--	
	1637	4.04x10 ⁶	1.41x10 ⁶	--	--	--	
	1735	4.71x10 ⁶	1.40x10 ⁶	--	--	--	
	2020	3.83x10 ⁶	1.39x10 ⁶	--	--	--	
	2132	3.88x10 ⁶	1.64x10 ⁶	--	--	--	
	2258	5.58x10 ⁶	1.60x10 ⁶	--	--	--	
3-8-57							Loop restored to reference conditions
3-9-57	0838	1.27x10 ⁷	1.32x10 ⁶	--	--	--	
3-10-57	2315	--	--	--	--	--	End of test

TABLE V

SUMMARY OF RESIN ACTIVITIES, WAPD-29 AND -2
RESIN ACTIVITY (total dis/min)

Nuclide	WAPD-29-1			WAPD-29-2			
	First Cycle Resin A	Second Cycle Resin E	Third Cycle Resin C	First Cycle Resin A	Resin B	Second Cycle Resin C	Resin D
I ¹³¹	4.99×10^{10}	3.53×10^{11}	6.82×10^{10}	3.28×10^{10}	5.51×10^{10}	4.02×10^{10}	5.47×10^9
Cs ¹³⁷	2.64×10^8	3.51×10^8	2.36×10^9	1.59×10^7	1.64×10^8	7.17×10^8	7.07×10^7
Sr ⁸⁹	2.97×10^9	4.85×10^9	2.64×10^9	8.35×10^7	1.46×10^9	2.32×10^8	1.67×10^9
Sr ⁹⁰	8.20×10^6	1.42×10^7					
Ba ¹⁴⁰	3.75×10^8	1.13×10^9	5.02×10^9	2.41×10^8	1.32×10^9	1.76×10^8	4.93×10^8
Zr ⁹⁵	1.36×10^8	8.53×10^7	2.07×10^8	1.78×10^8	3.91×10^8	2.44×10^8	3.02×10^7
Ce ¹⁴⁴	4.37×10^7	1.79×10^7	2.35×10^7	1.04×10^7	5.54×10^7	9.31×10^6	6.32×10^6
Mo ⁹⁹	--	9.56×10^5	--	--	--	--	--
Resin weight (g)	168	164	145	165	170	166	180
Time on stream (sec)	1.27×10^6	1.40×10^6	8.30×10^5	2.94×10^5	9.23×10^5	9.77×10^5	4.77×10^5
Date removed	12-16-56	1-6-57	1-28-57	2-5-57	2-18-57	3-4-57	3-10-57
Relative neutron flux level	High	High	Low	High	High	Low	High
Comments	Normal	Normal	Normal	Loop drain occurred	Normal	Normal	Oxygen accident studied

TABLE VI
ION EXCHANGE* DECONTAMINATION OF LOOP WATER

Loop Water Activities (dis/min-ml)

Date and Time	Cs^{138}		I^{131}		I^{133}		Ba^{139}	
	Before I X	After I X	Before I X	After I X	Before I X	After I X	Before I X	After I X
12-5 1000	8.10×10^6	$1.11 \times 10^{6**}$	9.64×10^4	N D†	8.30×10^5	N D	1.21×10^5	N D
12-6 0825	7.23×10^6	3.91×10^5	9.26×10^4	N D	1.15×10^6	N D	1.32×10^5	6.69×10^1
12-7 1255	9.42×10^6	3.98×10^5	8.80×10^4	N D	8.40×10^5	N D	1.32×10^5	6.01×10^1
12-10 0949	7.68×10^6	2.18×10^5	4.33×10^4	N D	5.45×10^5	N D	--	--
12-13 1058	--	--	3.63×10^4	N D	3.25×10^5	N D	--	--
12-14 1421	--	--	2.25×10^4	N D	2.30×10^5	N D	--	--
1-16 1249	--	--	--	--	--	--	--	--
1-21 1041	--	--	--	--	--	--	--	--
1-22 0925	--	--	--	--	--	--	8.90×10^2	N D
1-24 0938	--	--	--	--	--	--	9.01×10^2	N D

* LiOH form resin used

** Not corrected for Cs^{138} growth from 17m Xe^{138} (all other values are corrected).

† N D indicates that sample counted was not significantly greater than background, hence, not detectable.

TABLE VI (Cont)

Cs^{139}		Cs^{137}		Ba^{140}		Np^{239}	
Before I X	After I X	Before I X	After I X	Before I X	After I X	Before I X	After I X
--	--	6.61×10^1	6.61	3.69×10^1	1.17×10^1	--	--
--	--	2.26×10^1	2.78	--	--	--	--
--	--	--	--	--	--	--	--
--	--	--	--	--	--	--	--
--	--	2.53×10^1	7.92	--	--	--	--
--	--	--	--	--	--	3.21×10^3	2.82×10^2
7.98×10^3	3.47×10^3	--	--	--	--	--	--
7.02×10^3	N D	--	--	--	--	--	--
8.93×10^3	N D	--	--	--	--	--	--
--	--	--	--	--	--	--	--

where

- ν^a = probability per unit time that an atom will escape from the defected fuel elements (sec^{-1}),
 R = rate of escape of an isotope escaping from the fuel to the external system (atoms/sec),
 $(N^a)_f$ = total amount of the isotope in the fuel element (atoms).

The explicit equations used to calculate values of ν can be found in Ref 4. Variables used in the calculation of ν for the individual ion exchange resin beds during both tests are presented in Table II. Fissioning rates used in these calculations were obtained from heat fluxes which were determined from calorimetric measurements. That is, loop water temperatures observed before and after the in-pile test section were used in conjunction with flux plots and specimen locations to calculate individual specimen heat fluxes.

Calculated values of ν for both resin and loop water samples for WAPD-29-1 are given in Table VII. Table VIII summarizes the escape rate coefficient ν calculated for both resin and loop water samples for WAPD-29-2.

TABLE VII
WAPD-29-1 ESCAPE RATE COEFFICIENT ν CALCULATED FROM
PURIFICATION RESIN AND LOOP WATER ACTIVITIES (sec^{-1})

	Time Interval					
	Resin A on Stream		Resin B on Stream		Resin C on Stream	
	Resin	Loop Water	Resin	Loop Water	Resin	Loop Water
Xe ¹³⁸	--	1.78×10^{-7} (2)*	--	--	--	--
Kr ⁸⁸	--	3.38×10^{-7} (4)	--	1.06×10^{-7} (1)	--	1.27×10^{-8} (4)
I ¹³¹	6.08×10^{-10}	1.90×10^{-9} (6)	1.78×10^{-9}	--	6.14×10^{-10}	1.63×10^{-9} (3)
I ¹³³	--	3.83×10^{-9} (6)	--	--	--	2.21×10^{-9} (2)
Cs ¹³⁷	1.23×10^{-9}	--	4.08×10^{-10}	--	2.68×10^{-9}	--
Cs ¹³⁸	--	2.23×10^{-7} (8)	--	1.63×10^{-7} (1)	--	9.42×10^{-9} (9)
Cs ¹³⁹	--	--	--	2.67×10^{-8} (2)	--	7.19×10^{-9} (6)
Sr ⁸⁹	8.37×10^{-11}	--	4.88×10^{-11}	--	2.23×10^{-11}	--
Sr ⁹⁰	3.40×10^{-11}	--	1.23×10^{-11}	--	--	--
Ba ¹⁴⁰	2.75×10^{-12}	8.84×10^{-13} (2)	1.73×10^{-12}	--	1.90×10^{-11}	--
Zr ⁹⁵	3.85×10^{-12}	--	5.52×10^{-13}	--	1.79×10^{-12}	--
Ce ¹⁴⁴	5.60×10^{-12}	--	4.85×10^{-13}	--	7.61×10^{-13}	--
Mo ⁹⁹	--	--	4.42×10^{-11}	--	--	--
Relative neutron flux level	High		High		Low	

* Numbers in parentheses indicate number of determinations included in the calculation.

TABLE VIII
WAPD 29-2 ESCAPE RATE COEFFICIENT ν CALCULATED FROM
PURIFICATION RESIN AND LOOP WATER ACTIVITIES (sec⁻¹)

	Time Interval							
	Resin A		Resin B		Resin C		Resin D	
	Resin	Loop Water	Resin	Loop Water	Resin	Loop Water	Resin	Loop Water
Kr ⁸⁸	--	--	--	4.65×10^{-8} (2)*	--	7.55×10^{-9} (2)	--	2.63×10^{-8} (10)
I ¹³¹	2.24×10^{-9}	--	5.78×10^{-10}	2.93×10^{-10} (2)	4.24×10^{-10}	--	8.80×10^{-11}	--
Cs ¹³⁷	5.88×10^{-10}	--	3.57×10^{-10}	--	8.90×10^{-10}	--	1.50×10^{-10}	--
Cs ¹³⁸	--	1.28×10^{-7} (2)	--	6.58×10^{-8} (4)	--	2.59×10^{-8} (3)	--	3.09×10^{-8} (10)
Cs ¹³⁹	--	2.38×10^{-8} (1)	--	3.15×10^{-8} (2)	--	--	--	--
Sr ⁸⁹	1.54×10^{-11}	--	3.10×10^{-11}	--	2.38×10^{-12}	--	3.01×10^{-11}	--
Ba ¹⁴⁰	1.29×10^{-11}	--	5.97×10^{-12}	--	7.27×10^{-13}	--	3.59×10^{-12}	--
Ce ¹⁴⁴	1.29×10^{-11}	--	4.11×10^{-12}	--	4.00×10^{-13}	--	4.68×10^{-13}	--
Zr ⁹⁵	4.66×10^{-11}	--	6.44×10^{-12}	--	2.52×10^{-12}	--	4.05×10^{-13}	--
Relative neutron flux level	High		High		Low		High	
Comments:	Loop drain occurred		Normal		Normal		Oxygen accident studied	

* Numbers in parentheses indicate number of determinations included in the calculation.

TABLE IX

SUMMARY OF ION EXCHANGE PERFORMANCE

Nuclide	Ave Influent Activity (dis/min-ml)	Ave Influent Activity (dis/min-ml)	No. of Deter- minations (Ave)	Average DF*	Demineralizer Efficiency (%)**
Cs ¹³⁸	8.11×10^6	3.36×10^5	3	24.1	95.9
I ¹³¹	6.32×10^4	(10)†	6	$\geq 6.32 \times 10^3$	99.9
I ¹³³	6.53×10^5	(10)†	6	$\geq 6.53 \times 10^4$	99.9
Ba ¹³⁹	1.32×10^5	63.5	2	2.08×10^3	99.9
Cs ¹³⁷	3.82×10^1	5.77	3	6.6	84.9
Cs ¹³⁹	7.98×10^3	3.47×10^3	1	2.3	56.5
Ba ¹⁴⁰	3.69×10^1	1.17×10^1	1	3.2	68.3
Np ²³⁹	3.21×10^3	2.82×10^2	1	11.4	91.2

* $\frac{\text{Influent Activity}}{\text{Effluent Activity}} = \text{DF (Decontamination Factor)}$

** $\frac{\text{Influent Activity} - \text{Effluent Activity}}{\text{Influent Activity}} \times 100 = \text{Demineralizer Efficiency (\%)}$

† Assumed value (see text below)

Table IX presents a summary of purification resin decontamination factors for the various radio-nuclides. These calculations were based on data presented in Table V. The decontamination factor is evaluated by dividing the activity before the demineralizer filter by the activity after the demineralizer. It may be noted that the I¹³¹ and I¹³³ activities determined in the effluent samples were reported as "not detectable". It is felt, however, that, considering the effluent sample volume which was 10 cc and taking into account counting statistics, a figure of 10 dis/min-ml may be regarded as the lower limit of detectability. Hence, decontamination factors are calculated on this basis and expressed on a "greater than" basis. The decontamination factors for isotopes having short-lived, rare gas precursors may be low because of the daughter activity growth from a parent which is not removed by the demineralizer.

VII. CONCLUSIONS AND INTERPRETATION OF RESULTS

A. Loop Water Chemistry

The loop water chemistry conditions were maintained at predetermined levels of oxygen and hydrogen concentration, pH, and conductivity. During the "oxygen accident" phase of the test, the pH was 9.4 after oxygen at a concentration of 17.4 ppm was introduced into the loop water. This undoubtedly was a result of the chemical combination of dissolved oxygen with nitrogen under high irradiation to form oxides of nitrogen which, in turn, lowered the pH. However, the LiOH-form resin in the side stream purification system functioned well and essentially maintained the high pH loop water conditions.

B. Radiochemical Observations

A comparison of WAPD-29-1 and WAPD-29-2 escape rate coefficients computed from resin data is given in Table X. The comparison was made by normalizing the values of ν for a given isotope. That is, the values listed are based upon the lowest value of ν for the isotope within the two test groups.

Using this minimum value as a common denominator, the rest of the figures for the inter-test comparison were calculated. Therefore, the base number will be listed as 1.0, and all other numbers are multiples of this base figure. The values calculated for WAPD-29-1 (low density fuel specimens) are, in general, slightly higher than those calculated for WAPD-29-2 (high density fuel specimens). However, when the test average values are inspected there is great variability. This is especially true for the isotopes I^{131} , Cs^{137} , Sr^{89} , and Ba^{140} . Two of the radionuclides studied, namely Zr^{95} and Ce^{144} , indicated an opposite trend. The average test values for these isotopes were higher for the WAPD-29-2 experiment than for WAPD-29-1. Thus, this presents conflicting evidence as to whether there is a difference of fission product release from defected fuel specimens containing UO_2 fuel material of two different specific densities. Table XI is a comparison of WAPD-29-1 and WAPD-29-2 escape rate coefficients computed from loop water activities. By comparing the test averages, the values computed for WAPD-29-1 appear to be higher than those for WAPD-29-2. However, when the individual values are inspected, there is again as much variation within each test as there is difference between tests. From Tables IX and X, comparisons can be made between pairs of ν values for the same isotope to determine whether a consistent trend exists with time. Considering only the data obtained in the high flux positions, there are 26 such pairs of data; in all but three (I^{131} for periods A and B, 29-1; Sr^{89} for A and B, 29-2; and Cs^{139} for A and B, 29-2), ν decreases with time.

The values of ν for Kr^{88} and Cs^{138} calculated for both the WAPD-29-1 and WAPD-29-2 tests decreased markedly when the specimens were moved from the high neutron flux to the low flux region of the reactor.

In the WAPD-29-1 test, the values of ν for Kr^{88} and Cs^{138} decreased by factors of 13 and 20, respectively, when the flux was lowered. In the WAPD-29-2 test, the average values of ν for Kr^{88} and Cs^{138} were depressed by factors of 5 and 8, respectively. This effect is to be expected since ν should vary with fuel temperature to the extent that the diffusion mechanism is governed by an energy of activation. Since the escape rate coefficient ν is related to the diffusion coefficient (Ref 5), it would be expected that a change in heat flux resulting in a change in fuel temperature should change the value of ν . Although attempts have been made to confirm this effect in previous irradiation tests, results have been inconclusive to date.

The values of the escape rate coefficient are compared with those from the X-1-d test in Table XII. It can be seen that the "best" values of ν calculated for I^{131} and Cs^{137} from resin data of the X-1-d test were higher than those average values calculated for WAPD-29-1 and -2 by factors of 11 and 13, respectively. Values calculated for Sr^{89} , Sr^{90} , Ba^{140} , Zr^{95} , and Ce^{144} were not significantly different from those of the X-1-d test. Values for Kr^{88} were greater than those in the reference test. It may be pointed out that the third PWR fuel material test, X-1-d, which was run in the NRX Reactor (AECL) was considered a reference or "normal test" because it was the first test in which a near reference PWR rod was irradiated. The lower values of I^{131} and Cs^{137} may be the result of a basic difference in the operating characteristics of the MTR. Normally, the MTR maintains a constant power during a reactor cycle, while the NRX reactor in which the X-1-d test was run ordinarily has many power transients during a reactor cycle. It is more than likely that an increased rate of evolution during such power transients may have the effect of increasing the accumulation of activity on the ion exchange resin which is normally assumed to be a steady-state accumulation. Waterlogging may be the mechanism by which soluble nuclides escape to the loop water from defected elements in greater quantity than normal. Waterlogging, which is discussed in detail in Ref 4, may be defined as an activity transport mechanism which is postulated to occur when there are fuel temperature changes causing coolant water to be drawn into a defected specimen. When power is increased the coolant water is expelled as steam which carries leached fission products. It is expected that the coolant activity of readily leachable long-lived fission products would be most affected by this mechanism.

TABLE X

RELATIVE ESCAPE RATE COEFFICIENTS γ CALCULATED FROM FESIN DATA

WAPD-29-1 Low Density Fuel Specimens

WAPD-29-2 High Density Fuel Specimens

Nuclide	Relative Flux Levels				Relative Flux Levels				
	Resin A High	Resin B High	Resin C Low	Test Average	Resin A High	Resin B High	Resin C Low	Resin D High	Test Average
I ¹³¹	6.9	20.2	7.0	11.3	25.4	6.6	4.8	1.0	9.5
Cs ¹³⁷	8.2	2.7	17.9	9.6	3.9	2.4	5.9	1.0	3.3
Sr ⁸⁹	35.2	20.5	9.4	21.7	6.5	13.0	1.0	1.3	5.5
Sr ⁹⁰	2.8	1.0	--	--	--	--	--	--	--
Ba ¹⁴⁰	3.8	2.4	26.1	10.8	17.8	8.2	1.0	4.9	8.0
Zr ⁹⁵	9.5	1.4	4.4	5.1	115.1	15.9	6.2	1.0	34.6
Ce ¹⁴⁴	14.0	1.2	1.9	5.7	32.2	10.3	1.0	1.2	11.2

TABLE XI

RELATIVE ESCAPE RATE COEFFICIENTS γ CALCULATED FROM LOOP WATER ACTIVITIES

WAPD-29-1 Low Density Fuel Specimens

WAPD-29-2 High Density Fuel Specimens

Nuclide	Relative Flux Levels				Relative Flux Levels				
	Resin A High	Resin E High	Resin C Low	Test Average	Resin A High	Resin B High	Resin C Low	Resin D High	Test Average
Kr ⁸⁸	31.6	14.1	1.7	15.3	--	6.2	1.0	3.5	3.6
I ¹³¹	21.6	--	18.5	20.2	--	3.3	--	--	3.3
I ¹³³	3.2	--	1.6	2.4	--	--	--	--	--
Cs ¹³⁸	23.7	17.3	1.0	14.0	13.6	7.0	2.8	3.3	6.7
Cs ¹³⁹	--	3.7	1.0	2.4	3.3	4.4	--	--	3.9
Ba ¹³⁹	10.4	5.7	1.0	5.7	--	--	--	--	--

TABLE XII

ESCAPE RATE COEFFICIENTS CALCULATED FROM THE WAPD-29-1 AND -2
COMPARED WITH VALUES OF THE X-1-d ("NORMAL") TEST

		ν , sec ⁻¹		
		X-1-d	WAPD-29-1	WAPD-29-2
UO ₂ Density:		95.2	93.5	97.4
Isotope	"Best" Values*	Experimental Ave		
Kr ⁸⁸	1.8×10^{-8}	1.8×10^{-8}	1.2×10^{-7}	2.7×10^{-8}
I ¹³¹	1.0×10^{-8}	2.9×10^{-9}	9.9×10^{-10}	8.4×10^{-10}
Cs ¹³⁷	1.2×10^{-8}	1.2×10^{-8}	1.4×10^{-9}	5.0×10^{-10}
Sr ⁸⁹	9×10^{-12}	4.7×10^{-12}	5.2×10^{-11}	1.3×10^{-11}
Sr ⁹⁰	1.3×10^{-11}	7.8×10^{-12}	2.3×10^{-11}	--
Ba ¹⁴⁰	8.4×10^{-12}	4.5×10^{-12}	7.9×10^{-12}	5.8×10^{-12}
Zr ⁹⁵	1.8×10^{-12}	1.8×10^{-12}	2.1×10^{-12}	1.4×10^{-11}
Ce ¹⁴⁴	1.4×10^{-12}	1.4×10^{-12}	2.3×10^{-12}	4.5×10^{-12}

* There were limited experimental results available which were highly variable. However, it is believed that these represent the best values of calculation and comparison purposes.

TABLE XIII

COMPARISON OF ACTIVITIES AND VALUES OF ν DURING "OXYGEN ACCIDENT" PHASE OF TEST

Phase	Date	Kr ⁸⁸		Cs ¹³⁸	
		Ave (dis/min-ml) $\times 10^{-6}$	ν , sec ⁻¹ $\times 10^8$	Ave (dis/min-ml) $\times 10^{-6}$	ν , sec ⁻¹ $\times 10^8$
Before O ₂ Addition	3-5 and 3-6	2.80 ± 0.42 (5)*	1.6	1.24 ± 0.13 (5)	2.8
During period of high O ₂ concentration	3-7	4.19 ± 0.81 (12)	2.3	1.44 ± 0.16 (12)	3.3
After O ₂ removed and coolant restored to reference conditions	3-9	12.70 (2)	7.1	1.32 (2)	3.0

* Numbers in parentheses represent the number of determinations included in the calculations.

Table XIII lists the loop water specific activities and values of ν for Kr^{88} and Cs^{138} for the period prior to, during, and after the oxygen addition to the loop coolant. Apparently there was no significant effect on the rate of fission product evolution from the defected rod when it was exposed for a comparatively short time to a high oxygen concentration. The 32-hr exposure of the defected rod to an average oxygen concentration of 12.4 cc/kg appeared not to alter either the radionuclide activity levels observed in loop water and/or the activity levels analyzed on resin D which was on stream during this phase of the test. Escape rate coefficients for resin D follow the general downward trend with time described previously.

The ion exchange resin decontamination factors of I^{131} and I^{133} (Table IX) and the apparent decontamination factor of Ba^{139} are much greater than those of Cs^{137} , Cs^{138} , Ba^{140} , and Np^{239} . However, even the lowest decontamination factor, 2.3, which was computed for Cs^{139} , indicates a demineralizer removal efficiency of 56.5%. The apparent low decontamination factor of Cs^{139} may be a reflection of the decay of its rare gas parent Xe^{139} which has a 41-sec half-life. The growth of Cs^{139} and other cesium isotopes from their respective rare gas parents would result in poor apparent demineralizer efficiencies and decontamination factors. The demineralizer efficiency calculated in the normal manner may be computed by subtracting the effluent activity from the influent activity and dividing by the effluent activity. Thus, the actual amount of activity removed per pass through the sidestream demineralizer may be computed. Compared with the decontamination factors, the calculated efficiencies are much less sensitive to wide activity fluctuations in either influent or effluent activities. It can be seen from Table X that the decontamination factors of I^{131} and I^{133} are high, while the decontamination factors of Ba^{140} and Cs^{137} are comparatively low. This is in agreement with the decontamination factors determined during the ninth PWR fuel materials test, X-1-L, as reported in Ref 7. This, however, was only to be expected since the same type of resin was used in both experiments under essentially the same chemical conditions and flowrates.

ACKNOWLEDGMENTS

Radiochemical data at the reactor site were obtained by L. D. McIsaac and J. C. Teets. S. L. Mitchell, Jr. supervised the engineering aspects of the test at the reactor site. The author wishes to thank F. P. Mrazik, and F. Susko for supplying operational data and post-irradiation interpretations used in the preparation of this report.

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APPENDIX: TABLE A-I, CHEMISTRY OF LOOP WATER

Date	Time	Dissolved Gases (cc/kg)			pH	Remarks
		H ₂	Inerts	Total		
2-1-57	--	--	--	--	--	Start of WAPD-29-2 Test
2-22-57	1400	--	--	--	nil	10.4
2-25-57	1400	--	--	--	nil	10.4
2-26-57	1000	--	--	--	nil	10.3
	1430	30.1	9.7	39.8	--	--
						Injected 50 cc H ₂ at 1100 psig
2-27-57	1030	--	--	--	nil	10.2
2-28-57	1330	36.3	15.7	51.0	nil	10.2
3-1-57	1330	--	--	--	nil	10.1
3-4-57	1530	--	--	--	nil	10.1
3-5-57	1330	27.9	12.3	40.2	nil	10.1
3-7-57	0020	--	--	--	nil	10.1
						Injected O ₂ to give 41 cc/kg 27 cc/kg O ₂ by standard Winkler Analysis
	0130	0.0	0.0	--	10.2	--
	0230	--	--	--	9.7	9.4
	0300	--	--	--	20.3	9.3
	0400	0.0	0.0	--	11.5	--
	0500	--	--	--	24.7	--
	0530	--	--	--	23.6	9.3
	0700	0.0	0.0	--	22.1	--
	0800	--	--	--	19.9	9.3
	0930	--	--	--	13.2	--
	1100	--	--	--	12.5	--
	1300	--	--	--	11.7	--
	1330	0.0	0.0	5.1	--	--
	1550	--	--	--	--	--
	1530	--	--	--	19.3	--
	2055	--	--	--	26.2	--
	2315	--	--	--	19.4	--
						Injected O ₂ to give 20 cc/kg