MASTER

Argonne National Laboratory

TESTS OF DEFECTED THORIA-URANIA
FUEL SPECIMENS IN EBWR

by

R. F. S. Robertson

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FUEL SPECIMENS IN EBWR

by

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ABSTRACT

A potential thermal reactor fuel is provided by ThO_2 enriched with U^{235}O_2 . The two tests described in this report were carried out in EBWR to establish the rates of release of fission products, especially gases, from such a fuel and to assess the magnitude of the resulting operational problems.

The fuel used in both tests was 90 wt % ThO_2 , 10 wt % U^{235}O_2 in the form of pellets. In Test No. 1 the average diameter of the pellets was 0.300 in., and five pellets made a total fuel length of 3.1 in. The average density was 9.5 g/cc. The pellets were sheathed in a stainless steel tube welded shut at both ends; a 0.010-in. diameter hole was drilled through the sheath at the center of the fuel rod. In Test No. 2 a much larger fuel assembly was used; 82 pellets of 0.312-in. diameter made a total fuel length of 48 in., i.e., the same length as the fuel in an EBWR fuel plate. Average density was 9.0 g/cc. Again both ends were welded shut and the sheath was defected with a 0.20-in. diameter hole located at the top of the top pellet of the assembly.

The fuel specimen was mounted in a coolant channel of a natural uranium fuel element and was irradiated in EBWR. In each test the maximum thermal neutron flux in the fuel (using $\sigma_f = 545$ barns) was about 8×10^{12} n/(cm²)(sec). Surface heat rates were about 160,000 Btu/(hr)(ft²) (max) in both tests. Test 1 continued for a period corresponding to about 1.2% burnup of the U^{235} atoms while the second test had a burnup of about 3.2% (max) of the U^{235} atoms.

During each test, activities of various nuclides in the reactor water were measured, but most emphasis was placed on measurement of the rate of evolution of fission product gases. This was done by taking samples of air ejector gases and analyzing, by chemical means and by use of a 200-channel γ -ray spectrum analyzer, for the four nuclides Xe^{138} , Kr^{88} , Xe^{135} , and Xe^{133} . From the measured activity in the sample, knowing the flow rate of the air ejector gases, the evolution rates of these nuclides were calculated. Evolution rates were also measured as a function of reactor power.

During both tests no increase in the gross activity of the reactor water could be observed. This is not surprising in view of the fact that during normal operation the major activities in the water were due to the activation of corrosion products and structural materials rather than to the presence of fission products.

Radiation-monitoring instruments in the steam lines or in the air ejector gases did not show any significant increase in counting rate during either test. There is, however, an extremely high background due to N^{16} , which completely overshadows any activity due to fission product gases. Analyses of the air ejector gases did show that evolution rates of fission product gases had increased over those observed when no defect was present.

It was found that if the control rod nearest to the defected specimen was moved so as to increase the neutron flux in that specimen, a burst of activity was emitted; this was indicated by a peak in the counting rate of the γ -ray monitor at the air ejectors. This phenomenon was not observable when any other control rod was moved.

It was found that for fission product gases, the quotient R/Y was directly proportional to $1/\sqrt{\lambda}$, where R was the rate at which gas was escaping from the defect (atoms/sec), Y was the fission yield, and λ (sec^{-1}) was the decay constant of the gas. It can be shown that this relationship is consistent with release of gases by a diffusion-controlled mechanism through the oxide.

1. INTRODUCTION

Ceramic fuels are continually gaining popularity for use in thermal reactors. Two such fuels used to date have been uranium oxide (UO_2) enriched in U^{235} , and thorium oxide-uranium oxide ($\text{ThO}_2\text{-UO}_2$) enriched in U^{235} . A great deal of information is available on the irradiation behavior of UO_2 ; ^(1,2,3) however, very little is known about the irradiation behavior of $\text{ThO}_2\text{-UO}_2$, especially regarding the release of fission products from a defective fuel element. To study this latter question and to gain a knowledge of the distribution of fission products through the system, a series of tests with defective $\text{ThO}_2\text{-UO}_2$ fuel specimens were conducted in the Experimental Boiling Water Reactor (EBWR).

The EBWR is a natural circulation, direct-cycle boiling water reactor fueled with plates of enriched uranium-zirconium-niobium alloy sheathed in Zircaloy-2.^(4,5) During these tests the reactor was used merely as an irradiation facility. This report deals only with the behavior of defective ceramic fuel elements in a boiling water reactor and makes no reference to uranium alloy fuels.

2. OBJECTIVES

The three main objectives of these tests were:

- (1) To study the rate of release of fission products from a defective thorium-uranium fuel element.
- (2) To study the distribution of these fission products throughout the reactor system.
- (3) To attempt to assess the hazards involved in operating a boiling water reactor fueled with a ceramic fuel which contains defects in the cladding.

3. EXPERIMENTAL METHODS

Two tests were performed with defective thorium-uranium fuel specimens. The first test, referred to hereafter as Test 1, was on a small specimen and was run mainly to determine the order of magnitude of activity levels which would be involved. The second test was on a much larger specimen. The dates of these tests were:

Test 1	April 24 to July 11, 1958
Test 2	July 21 to November 7, 1958.

3.1 Fuel Specimens

The thorium-uranium ceramic used was in the form of pellets manufactured by the Ceramics Group, Metallurgy Division, Argonne National Laboratory. These pellets were prepared by cold pressing a $\text{ThO}_2\text{-U}_3\text{O}_8$ mixture and then firing the pellets in air at 1750°C . Some details regarding composition, enrichment, density, etc., may be found in Table 3-1.

Before acceptance, a few pellets were tested by autoclaving in distilled water at 680°F for 65 hr. No significant weight gains were observed.

The sizes of stainless steel tubing were chosen such that the diametral clearances were as shown in Table 3-1. The end clearance between the bottom of the top plug and the top of the top pellet was about $\frac{1}{8}$ in. A hole of the size and location indicated in Table 3-1 was then drilled through the stainless steel sheath.

The stainless steel tubes into which the pellets were loaded were prepared as follows: A plug was first welded into the bottom of the tube. This was tested for absence of leaks by pressurizing with helium and testing outside the tube with a helium leak detector. The required number of pellets were then loaded into the tubes and the top plug was welded in place.

Table 3-1

TEST SPECIMENS

	Test 1	Test 2
Composition, wt%		
UO ₂ .1	10	10
ThO ₂	90	90
Density		
gm/cc	9.7	9.1
% Theoretical	95	90
U ²³⁵ Enrichment, wt % U	93	93
Wt U ²³⁵ , gm	2.73	43.5
Pellet Diameter, in.	0.300 ± 0.001	0.312 ± 0.002
Fuel Length, in.	3.1	48
Fuel Sheath Clearances (cold), in.		
Diametral	0.004	0.010
Axial	0.125	0.125
Sheath Material	Stainless Steel	
Sheath Thickness, in.	0.035	0.025
Sheath Defect		
Location	Center of Fuel	Top of Fuel
Diameter, in.	0.010	0.020

FIG. 3-1

SCHEMATIC INDICATION OF METHOD OF ATTACHING FUEL SPECIMENS TO EBWR FUEL ELEMENT AND LOCATION OF FUEL ELEMENT IN EBWR.

Stainless steel, $\frac{3}{8}$ -in. diameter rod was welded to the top and bottom plugs and the assembly was then fitted in a "natural-thin" EBWR fuel element,⁽⁴⁾ as indicated in Fig. 3-1. The bottom length of the rod fitted into a recess in the lower locating end fitting of the fuel element, while the top end was fastened to the stainless steel top fixture of the fuel element by a screw.

In the two tests the fuel specimens were located in the fuel element as follows:

Test 1: Centerline of specimen at point of maximum thermal neutron flux.

Test 2: Top and bottom of specimen level with top and bottom of meat in EBWR fuel plate.

The slot shown in Fig. 3-1 was provided as a starting point for the hacksaw blade used for cutting out the fuel specimen from the fuel element subsequent to irradiation.

Figure 3-1 also shows a schematic plan view of the EBWR fuel element containing the fuel specimen. The location of the fuel element in the reactor is also indicated. Both tests were performed with the fuel specimen in the same location in the reactor.

3.2 Reactor Operation

A brief description will be given of EBWR operation, with reference only to particulars which are of interest to this test.

Figure 3-2 shows a schematic diagram of the various flow systems.

FIG. 3-2
SIMPLIFIED DIAGRAM OF FLOW SYSTEMS IN EBWR

Water is boiled in the reactor vessel and the steam is passed to the turbine. Beyond the turbine the steam is condensed in the condenser, and the condensate is then pumped from the condenser hotwell back to the reactor, passing through a filter enroute.

The condenser is maintained at an absolute pressure of about 2 in. Hg by removing noncondensable gases via the air ejectors, which are aspirators operated by steam. The feedwater merely acts as coolant to condense the aspirator steam. The noncondensable gases include H_2 and O_2 from radiolysis in the reactor and air which leaks into the condenser. These act as carrier gases for any fission product gases which may be stripped from the reactor water.

After passing the air ejectors, the gases are passed through a unit to recombine H_2 and O_2 . The noncondensable gases from inleakage into the condenser and fission product gases are passed through a filter to the blower which sends them out the reactor stack.

Purity of the water in the reactor vessel is maintained by passing a small fraction through the ion exchange system. The ion exchanger used is a mixed bed strong acid, strong base-type resin (MB-1) (Rohm and Haas).

Samples of reactor water, steam, feedwater, air ejector gases, or water leaving the ion exchanger are taken at sampling points which are shown in Fig. 3-2.

Table 3-2 shows EBWR normal operating conditions at a power of 20 Mw(t). Several parameters which will be useful in future calculations are included.

3.3 Irradiation

Figures 3-3a and 3-3b show the reactor power through the two tests.

Table 3-3 shows the operating characteristics of the two specimens.

Thermal neutron fluxes were obtained by analysis of the irradiation oxide pellets for Cs^{137} and SR^{90} . Details of the analyses and methods of calculation are outlined in Appendix 1. The fluxes quoted are those associated with a fission cross section of 545 barns for U^{235} , obtained from Westcott.⁽⁶⁾

For Test 2 the ratio of the maximum to average thermal neutron flux in the specimen was taken as 1.72.

Table 3-2

EBWR OPERATING CHARACTERISTICS AT 20 Mw (THERMAL)

	English System	Metric System
<u>Temperatures:</u>		
Reactor Water	489°F	254°C
Steam	489°F	254°C
Feedwater	120°F	49°C
Entrance to Ion Exchange	110°F	43°C
<u>Pressures:</u>		
Steam	600 psig	41.8 atm
Condenser	~1½ in. Hg abs	~40 mm Hg
Sample Station for Air Ejector Gases	12 in. H ₂ O	784 mm Hg
<u>Flow Rates:</u>		
Steam	60,000 lb/hr	7.56 kg/sec
(at 489°F)	740 ft ³ /min	349 l/sec
Feedwater	60,000 lb/hr	7.56 kg/sec
Water to Ion Exchanger	~10 gal/min	~0.63 kg/sec
<u>Gas Flow Rate at Sample Station:</u>		
Recombiner on	~1 SCFM	~28 l/min (STP)
Recombiner off	~2 SCFM	~57 l/min (STP)
<u>Volumes:</u>		
Recirculating Water (cold)	3,000 gal	1.14 x 10 ⁴ l
Steam Space in Reactor Vessel	~300 ft ³	8.5 x 10 ³ l
Piping, Reactor to Turbine (including Steam Dryer)	65 ft ³	1.84 x 10 ³ l
<u>Residence Times of Noncondensable Gases:</u>		
In Reactor Steam Dome	25 sec	
In Pipe from Reactor to Turbine	5 sec	
In Turbine and Condenser	30 sec	
In Piping from Air Ejectors to Stack Exit	90 sec

FIG. 3-3(a)

REACTOR POWER FOR TEST NO. 1
NUMBERS IN PARENTHESES
SHOW Mw-hrs OF OPERATION
FOR EACH POWER RUN

FIG. 3-3(b)

REACTOR POWER FOR TEST NO. 2

Table 3-3

OPERATING CHARACTERISTICS OF FUEL SPECIMENS;
REACTOR POWER 20 Mw (THERMAL)

	Test 1	Test 2	
		Maximum	Average
Reactor Operation, Mwd	787	2016.7	
nvt, n/cm ²	2.7×10^{19}	7.0×10^{19}	4.0×10^{19}
Burnup, at-% U ²³⁵	1.2	3.2	1.9
Thermal Neutron Flux, n/(cm ²)(sec) (See Appendix 1)	8×10^{12}	8×10^{12}	4.6×10^{12}
Fissions/sec	3.25×10^{13}	4.83×10^{14}	2.81×10^{14}
Surface Heat Flux (Fuel)			
watt/cm ²	52	48	28
Btu/(hr)(ft ²)	1.65×10^5	1.52×10^5	0.89×10^5
$\int k(T)dT$, watt/cm	10	9.6	5.6
Temperatures, °C (See Appendix 2)			
Central	900	1300	1000
Surface (Fuel)	650	1000	800

The calculation of fuel temperatures is outlined in Appendix 2.

3.4 Postirradiation Examination of Fuel

After irradiation, the fuel element containing the defect specimen was transferred from the reactor to the fuel storage pit. There the specimen was removed from the fuel element by cutting at the slot marked in Fig. 3-1. The specimen was then transferred in a lead flask to the hot-cell facilities of ANL, where it was sectioned and photographed.

3.5 Chemical and Radiochemical Analyses

3.5.1 Fission Product Gases

The release rate of fission gases was determined by taking samples of the exhaust gases from the air ejectors at Sample Station 6.

When each sample was taken, the flow rates of the exhaust gases were measured. These samples were then analyzed for individual nuclides either by chemical means or by γ -ray spectrometry using a 200-channel γ -ray pulse height analyzer.

Full details regarding sampling and both types of analyses may be found in Appendix 3.

The release rate for a given nuclide from the fuel defect was then calculated as follows:

$$\text{Curies/day} = 6.49 \times 10^{-7} A f e^{+\lambda \tau}$$

$$\text{Atoms/sec} = (A f / 3.6 \lambda) e^{+\lambda \tau}$$

where

A = Specific activity of nuclide in gas, disintegrations/(ml)(min)

f = Flow rate of exhaust gases, liters/min

λ = Decay constant of nuclide, sec^{-1}

τ = Time it takes gas atom to travel from reactor core to sampling station in air ejector gas stream, sec

Actually, for all fission gas nuclides determined the time was short compared to the half-life of the nuclide; hence the exponential in both the above expressions was unity to a very good approximation.

During Test 1, only release rates of Xe^{138} and Kr^{88} were measured. During the first part of Test 2 again only Xe^{138} and Kr^{88} were determined experimentally, but after September 19, when the 200-channel γ -ray pulse height analyzer was calibrated and ready for use, data were obtained on Xe^{138} , Kr^{88} , $\text{Kr}^{85\text{m}}$, Xe^{135} , and Xe^{133} .

3.5.2 Fission Products in Reactor Water

During both tests, samples of reactor water were taken from Sample Station 2 and were analyzed by conventional radiochemical means for fission products. Before sampling, enough HCl or HNO_3 was added to the sampling bottle so that the final sample would be 1N in acid when the bottle was full.

3.5.3 Gross Beta-Gamma Count Rate

Samples of reactor water and the effluent from the ion exchange resins were taken throughout the test. A known volume was evaporated to dryness on a stainless steel counting tray. These samples were

then counted and the decay was followed periodically on a gas flow proportional beta counter.

3.6 Metal Specimens in Steam Line and in Condenser

During both tests samples of various metals were placed both in the steam line and in the condenser to determine both the extent to which corrosion took place and the extent to which fission products were deposited on their surfaces.

The specimens in the steam line which were representative of turbine construction materials were in the form of plates 2.4 by 1 by 0.06 in. They were mounted in a special holder and were inserted in the steam line just before the steam dryer. They were positioned such that the long axis was perpendicular to and the 1-in. axis was parallel to the direction of steam flow.

The materials used were:

(1) Structural Steel	ASTM A-113 SSC
(2) Steel	AISI-4140
(3) Stainless Steel	Type 403 (13% Cr)
(4) Steel	1% Cr-0.5% Mo
(5) Low Carbon Steel	
(6) Nonhardening Stainless Steel	13% Cr

The specimens in the condenser which were representative of condenser structural materials were in the form of plates $\frac{7}{8}$ by $1\frac{1}{2}$ by 0.06 in. They were attached to the top row of condenser tubes such that the long axis was parallel to the length of the tube and the flat face was perpendicular to the direction of steam flow.

Materials used were:

- (1) Stainless Steel, Type 304
- (2) Mild Steel
- (3) Copper
- (4) X-198 Aluminum containing 1% Nickel and 0.1% Titanium
- (5) X-8001 Aluminum-1% Nickel Alloy

At the completion of each test both sets of samples were removed and were counted in a 256-channel γ -ray pulse height analyzer.

3.7 Sampling Tubes and Monitoring System

Four stainless steel tubes were inserted through swagelock fittings in the head of the reactor vessel. Each tube was placed to withdraw water from a specific location, either from above the fuel element containing the defect specimen or from adjacent, or far-removed, fuel elements containing no defect.

Reactor water was passed through these tubes. The water was then cooled and passed either to a delayed-neutron monitor or to a cation ion-exchange bed. Beyond the bed the activity of the water could be monitored and measured by a NaI(Tl) scintillator connected to a photomultiplier tube and single-channel γ -ray pulse-height analyzer.

The first monitor, the delayed-neutron monitor, counted neutrons after thermalization in a tank of water by means of a BF_3 tube connected to a linear amplifier, discriminator, and count rate meter.

The second monitor, the "Iodine Monitor," operated on the principle that the cation bed would remove cationic and filterable activities from the water, leaving only rare gases and anionic species such as radioiodines. Gamma rays from the rare gases, which are of low energy, could be discriminated against by setting the discriminator level to count only pulses with energies greater than 900 kev, and thus the monitor should "see" only radioiodines or bromines in the reactor water.

4. EXPERIMENTAL RESULTS

4.1 General Plant Radiation Levels

During both tests, with one exception noted in Section 4.2, the radiation levels registered by various instruments throughout the system were no higher than those normally registered. These instruments included radiation monitors at the following locations: Turbine, Hotwell, Steam Dryer, Air Ejectors, and Filter in Off-Gas System.

4.2 Behavior of Air Ejector Monitor during Power Transients

Normally, when no defective fuel specimens were present the counting rate registered by the radiation monitor at the air ejectors rose smoothly to a new level when the reactor power was increased.

During Test 1 this monitor showed normal behavior. During Test 2, however, it was found that when reactor power was increased the counting rate shown by this instrument rose initially to a maximum value and then dropped over a period of a few minutes to a steady level. Figure 4-1(a) shows typical behavior during changes in reactor power. This peaking phenomenon was observed only when reactor power was increased. When power was dropped, the counting rate fell smoothly to a lower level. This effect was also observed with the monitors at the hotwell and on the filter in the off-gas system.

At constant reactor power, it was observed that movement of No. 3 control rod also had an effect on the count rate of the air ejector monitor. Figure 4-1(b) illustrates typical behavior. When No. 3 rod was inserted fully into the core and the remainder of the rods were raised as a bank to maintain reactor power, the count rate was not greatly affected. If, now, No. 3 rod were fully withdrawn, the counting rate shown by the monitor peaked as shown. Both the reactor power and the steady counting rate were higher. When No. 3 rod was then returned to bank position, there was no appreciable effect.

Next, if No. 3 rod was lowered into the core and No. 9 was simultaneously raised to maintain reactor power, there was no effect. When the positions of the two rods were interchanged, i.e., when No. 3 was raised and No. 9 was lowered, the peaking in the count rate was again observed.

It will be remembered (Fig. 3-1) that the sample was located in the vicinity of No. 3 control rod. In general, it was found that only if that rod were moved so as to increase the neutron flux in the specimen, a peak was observed in the counting rate. Any movement of any other control rod had no transient effect on the count rate of the air ejector monitor.

FIG. 4-1
TRANSIENT BEHAVIOR OF AIR EJECTOR MONITOR

4.3 Monitor Results

During Test 1, the presence of delayed neutrons was detected by the delayed-neutron monitor, but no significant difference could be found between the counting rate of water samples from the vicinity of the defect specimen, and that of water from any of the other three locations above the reactor core. Similarly, the iodine monitor showed no difference between the four samples.

During Test 2, the sensitivity of the delayed-neutron monitor was greatly increased by the use of several BF_3 tubes mounted in parallel. In this test again, however, no significantly high counting rate could be detected by either monitor in water from above the defect specimen when compared to water from elsewhere in the reactor core.

No transient behavior of either monitor such as that described in the preceding section, was found during reactor power increases or during movement of No. 3 control rod.

4.4 Release Rates of Fission Product Gases

4.4.1 No Defect

In order to provide background information, the air-ejector gases and the reactor water were sampled during periods of reactor operation when no defective specimens were present. During these periods it was known that the only fission products present would be from uranium contamination of the Zircaloy-2 cladding of the EBWR fuel.

Table 4-1 shows the results obtained.

4.4.2 Test 1

Table 4-2 shows results obtained during the first test.

It is now thought that during this early part of the program not enough care was taken in Kr^{88} analyses, so that the results are low - probably by as much as a factor of two.

It will be noticed that both Xe and Kr release rates fell steadily as the experiment progressed. The only explanation produced to date for this effect is that the 0.010-in. diameter hole through the sheath gradually plugged as the experiment progressed. This effect was not observed during the second test.

Tables 4-3 and 4-4 show results obtained during the second test.

The samples of July 30 and 31, labeled "No. 3 Rod Fully Out," in Table 4-3 were taken during the activity transient observed when No. 3 rod was pulled out of the core. It can be seen that both Xe^{138} and Kr^{88} activities were considerably higher than normal during these periods. These values are not necessarily at the peak of the transient curve and maximum values may have been higher than those recorded.

Figure 4-2 shows a plot of the data of Table 4-4. The short-lived fission gas release rates were reasonably constant at a given power level and rose and fell with power changes in the manner expected.

When reactor power was increased, the release rates of all fission gases showed the same transient behavior immediately after the power change that had been observed on the air ejector monitor. Figure 4-2 shows that the magnitude of the peak increased as the half-life of the nuclide increased.

Table 4-1

RELEASE RATE OF FISSION GASES - NO DEFECT

Date	Time	Reactor Power (Mw)	Gas Flow Rate (l/min)	Activity in Gas [d/(min)(ml)]					Release Rate (curie/day)				
				Xe ¹³⁸ x 10 ⁻⁴	Kr ⁸⁸ x 10 ⁻³	Kr ^{85m}	Xe ¹³⁵	Xe ¹³³	Xe ¹³⁸	Kr ⁸⁸	Kr ^{85m}	Xe ¹³⁵	Xe ¹³³
11-24	0910	20	35	4.44	1.96		1520	119	1.0	0.045		0.035	0.0027
11-28	1010	20	37	4.54	2.20	455	1410	112	1.1	0.053	0.011	0.034	0.0027
11-28	1008	20	36	4.08	2.15	494	1470	111	0.96	0.050	0.012	0.035	0.0026
12-4	1030	20	58	3.23	1.76	336	1000	80.4	1.2	0.066	0.013	0.038	0.0030
12-18	1347	20	29	-	2.59	541	1650	74.8*	-	0.049	0.010	0.031	0.0014*

*Sample taken a few hours after reactor startup. Xe¹³³ not yet at saturation.

Table 4-2

Xe¹³⁸ AND Kr⁸⁸ RELEASE RATES - TEST 1

Date	Time	Reactor Power (Mw)	Gas Flow Rate (ℓ/min)	Activity in Gas [d/(min)(ℓ)]		Production Rate (curie/day)	
				Xe ¹³⁸	Kr ⁸⁸	Xe ¹³⁸	Kr ⁸⁸
4-24	1400	5	71	2.62 x 10 ⁶		0.12	
4-24	1930	5	62	4.8 x 10 ⁶		0.19	
4-28	1022	10	65	4.3 x 10 ⁶		0.18	
4-28	1210	15	59	7.4 x 10 ⁶		0.28	
4-28	1547	20	62	3.3 x 10 ⁷		1.34	
4-29	1548	20	73	2.9 x 10 ⁷		1.38	
5-1	0925	20	65	2.6 x 10 ⁷	2.4 x 10 ⁶	1.10	0.10
6-11	0940	20.4	97	2.0 x 10 ⁷		1.26	
6-11	1455	21.2	97	2.6 x 10 ⁷		1.64	
6-11	1457	21.2	97	2.7 x 10 ⁷		1.70	
6-12	1033	20.0	97	1.9 x 10 ⁷	1.4 x 10 ⁶	1.20	0.09
6-12	1400	20.9	97	2.4 x 10 ⁷	1.8 x 10 ⁶	1.51	0.11
6-13	1008	20.4	70	2.0 x 10 ⁷	1.6 x 10 ⁶	0.91	0.07
6-19	1103	20.9	61	2.6 x 10 ⁷	2.3 x 10 ⁶	1.03	0.09
6-19	1348	21.4	61	2.2 x 10 ⁷	2.0 x 10 ⁶	0.87	0.08
6-25	0933	20.6	62	1.5 x 10 ⁷		0.61	
*6-26	1320	20.3	32	2.7 x 10 ⁷	2.3 x 10 ⁶	0.56	0.05
*6-26	1345	20.6	32	2.2 x 10 ⁷	2.3 x 10 ⁶	0.46	0.05
6-27	0837	20.6	77	1.6 x 10 ⁷	1.0 x 10 ⁶	0.80	0.05
6-27	0911	20.6	77	1.5 x 10 ⁷	7.5 x 10 ⁵	0.75	0.04
6-27	1237	20.4	77	1.4 x 10 ⁷	6.5 x 10 ⁵	0.70	0.03
6-27	1237	20.4	77	1.7 x 10 ⁷	9.0 x 10 ⁵	0.85	0.045
7-1	1125	10.3	37	6.6 x 10 ⁶	4.0 x 10 ⁵	0.16	0.009
7-1	1428	10.7	34	8.2 x 10 ⁶		0.18	
7-1	1700	11.8	48	7.7 x 10 ⁶		0.25	
7-2	1103	24.6	55	1.7 x 10 ⁷	8.5 x 10 ⁵	0.61	0.03
7-2	1330	24.6	51	2.1 x 10 ⁷	6.7 x 10 ⁵	0.69	0.02
7-2	1537	28.9	69	1.6 x 10 ⁷	9.8 x 10 ⁵	0.72	0.04
*7-7	0921	20.2	30	2.3 x 10 ⁷	1.2 x 10 ⁶	0.45	0.02
7-7	1550	18.9	50	1.6 x 10 ⁷		0.52	
7-8	0925	19.9	58	~1.2 x 10 ⁷	4.2 x 10 ⁵	~ 0.45	0.016
7-8	1411	20.5	58	~1.0 x 10 ⁷		~ 0.38	
7-8	1526	20.2	55	1.6 x 10 ⁷		0.57	
7-9	0944	20.4	58	1.1 x 10 ⁷	3.8 x 10 ⁵	0.42	0.014

*Recombiner in operation.

Table 4-3

RELEASE RATES OF FISSION GASES - TEST 2

Date	Time	Reactor Power (Mw)	Gas Flow Rate (ℓ/min)	Activity in Gas [d/(min)(ml)]		Release Rate (curie/day)		Remarks
				Xe ¹³⁸ x 10 ⁻⁴	Kr ⁸⁸ x 10 ⁻³	Xe ¹³⁸	Kr ⁸⁸	
7-22	0909	5.02	39	1.0		0.26		
7-22	0910	5.02	39	1.0		0.26		
7-22	1139	9.9	56	1.15		0.42		
7-22	1517	14.7	56	1.80		0.65		
7-23	1315	19.8	51	4.60	8.75	1.5	0.29	
7-24	1327	20.8	57	4.20	10.7	1.6	0.40	
7-25	1044	19.8	57	4.25		1.6		
7-26	1247	20.4	53	4.20		1.5		
7-27	1415	20.4	53	3.60	10.2	1.3	0.35	
7-28	0925	20.0	48	4.10		1.3		
7-28	1312	20.0	47	3.60		1.1		No. 3 rod fully in.
7-30	0907	20.0	55	4.65		1.7		
7-30	1346	20.0	56	26.0	140.0	9.4	5.1	No. 3 rod fully out.
7-31	0912	20.0	57	4.50		1.7		
8-7	0933	20.0	59	3.90		1.5		
8-8	0910	20.0	53	4.25		1.5		
8-8	1257	20.0	44	11.0		3.1		No. 3 rod out.
8-11	0940	20.6	50	6.15	9.6	2.0	0.31	
8-12	1258	20.6	50	5.0		1.6		
8-13	0923	20.0	56	4.15	10.0	1.5	0.36	
8-25	1016	20.4	62	3.70		1.5		
8-27	0940	20.1	56	4.10		1.5		
8-29	0922	20.0	56	3.60		1.3		
9-3	1015	19.8	58	3.80	11.0	1.4	0.42	
9-5	0927	19.3	53	4.70	11.0	1.6	0.38	
9-11	1425	20.0	63	3.70		1.5		
9-15	1111	20.0	54	4.50	13.0	1.6	0.45	

Table 4-4
RELEASE RATES OF FISSION GASES - TEST 2

Date	Time	Reactor Power (Mw)	Gas Flow Rate (l/min)	Activity in Gas [d/(min)(ml)]					Release Rate (curie/day)				
				Xe ¹³⁸ x 10 ⁻⁴	Kr ⁸⁸ x 10 ⁻⁴	Kr ^{85m} x 10 ⁻³	Xe ¹³⁵	Xe ¹³³	Xe ¹³⁸	Kr ⁸⁸	Kr ^{85m}	Xe ¹³⁵	Xe ¹³³
9-19	1035	21.3	53	3.96	1.15	4.7	3760	1330	1.4	0.40	0.18	0.13	0.046
9-22	1536	10.7	45	1.62	0.50	3.5	5820	4760	0.47	0.15	0.103	0.17	0.14
9-23	1036	10.4	38	2.18	0.45	3.9	5500	5610	0.54	0.11	0.096	0.14	0.14
9-23	1457	10.8	47	1.97	0.47	2.5	5260	4600	0.60	0.14	0.076	0.16	0.14
9-24	0949	12.7	42	2.95	0.67	3.6	5960	4020	0.81	0.18	0.098	0.16	0.11
9-24	1146	12.7	41	2.91	0.64	3.1	5090	3430	0.78	0.17	0.083	0.14	0.092
9-24	1440	12.7	43	2.90	0.61	3.4	5430	3220	0.81	0.17	0.095	0.15	0.090
9-25	1004	14.8	55	2.41	0.79	3.7	4330	2500	0.86	0.28	0.13	0.16	0.089
9-25	1340	14.9	52	2.25	0.70	3.5	4250	1910	0.76	0.24	0.12	0.14	0.065
9-25	1554	14.9	57	2.45	0.77	3.3	4280	1850	0.91	0.28	0.12	0.16	0.069
9-26	1027	17.3	58	2.99	1.02	4.0	4400	1740	1.1	0.38	0.15	0.17	0.066
9-26	1334	17.4	49	3.29	0.96	4.4	4800	1630	1.1	0.31	0.14	0.15	0.052
9-26	1604	17.4	49	3.29	1.05	4.2	4970	1660	1.1	0.33	0.13	0.16	0.053
9-27	1047	20.1	63	3.15	0.85	-	3390	1020	1.3	0.35	-	0.14	0.042
9-27	1050	20.1	62	2.88	0.97	-	3500	1000	1.2	0.39	-	0.14	0.040
9-30	0939	20.4	27	7.50	2.30	10.5	7680	2020	1.3	0.40	0.18	0.14	0.035
10-1	1043	20.5	29	5.60	2.62	9.5	6940	1800	1.1	0.49	0.18	0.13	0.034
10-2	0940	20.0	25	6.82	2.73	9.5	8080	2020	1.0	0.44	0.15	0.13	0.033
10-3	1004	20.0	26	6.75	2.92	10.4	7570	2290	1.1	0.49	0.18	0.13	0.039
10-6	0937	20.4	26	6.53	2.81	10.8	7720	2140	1.1	0.48	0.18	0.13	0.36
10-6	1123	23.2	26	9.10	5.54	22.3	17100	6950	1.4	0.87	0.38	0.27	0.11
10-6	1403	23.4	52	4.04	1.69	5.8	4590	2080	1.4	0.57	0.20	0.16	0.070
10-7	1056	23.3	26	-	3.55	11.2	8240	2540	-	0.60	0.19	0.14	0.043
10-8	1017	23.2	25	8.47	3.98	14.1	9850	2670	1.4	0.65	0.23	0.16	0.043
10-9	0952	23.4	27	-	-	12.6	8960	2480	-	-	0.22	0.16	0.044
10-10	0904	23.3	28	7.46	3.06	9.8	7200	2220	1.4	0.56	0.22	0.13	0.040
10-13	0904	23.5	26	8.12	2.40	14.9	9680	3040	1.4	0.41	0.25	0.16	0.051
10-13	1411	26.5	32	8.40	3.41	13.6	9120	3870	1.7	0.71	0.28	0.19	0.081
10-13	1539	26.6	27	8.68	3.40	14.0	9040	3720	1.5	0.60	0.25	0.16	0.065
10-14	0916	26.2	27	8.60	3.22	13.4	9360	3020	1.5	0.57	0.23	0.16	0.053
10-15	0950	26.3	32	8.20	3.08	11.9	8700	2830	1.7	0.64	0.25	0.18	0.059
10-16	1034	26.4	30	8.75	3.51	9.2	9280	3080	1.7	0.69	0.18	0.18	0.060
10-17	0941	26.6	30	7.90	3.02	11.8	7630	2600	1.5	0.59	0.23	0.15	0.051
10-17	1342	19.9	43	3.45	1.07	2.90	3610	1040	1.0	0.30	0.081	0.10	0.029
10-18	1050	21.1	42	4.53	2.21	8.5	7730	4930	1.2	0.60	0.23	0.21	0.13
10-20	1002	20.0	68	2.91	1.53	7.4	5960	3000	1.3	0.68	0.33	0.26	0.13
10-21	1027	20.6	23	7.80	4.72	22.0	18100	8220	1.2	0.71	0.33	0.27	0.12
10-22	0900	20.3	30	6.00	3.16	12.2	9640	4140	1.2	0.62	0.24	0.19	0.081
10-23	0852	20.8	30	5.42	2.62	10.5	8940	3450	1.1	0.51	0.20	0.17	0.067
10-24	0927	20.5	30	5.83	3.14	13.8	10100	3540	1.1	0.61	0.27	0.19	0.069
10-25	1349	20.4	29	5.87	4.49	12.1	11100	3960	1.1	-	0.23	0.21	0.075
10-26	1648	20.4	30	-	-	8.1	8480	2680	-	-	0.16	0.17	0.052
10-27	0950	20.4	30	5.77	2.67	9.4	8560	2790	1.1	0.52	0.18	0.17	0.054
10-28	1331	20.7	32	5.41	2.42	9.2	8080	2370	1.1	0.50	0.19	0.17	0.049
10-29	1007	20.8	32	6.09	2.94	9.7	8800	2620	1.3	0.61	0.20	0.18	0.055
10-30	1042	20.8	31	5.94	2.28	8.3	8240	2350	1.2	0.46	0.17	0.17	0.047
10-30	1243	10.7	29	2.53	1.76	1.5	3630	916	0.48	0.33	0.03	0.07	0.017
10-31	0938	11.5	28	3.31	1.12	6.1	10800	8950	0.60	0.20	0.11	0.20	0.16
11-1	1005	12.0	27	3.01	1.65	7.7	12100	11400	0.52	0.29	0.13	0.21	0.20
11-2	1537	12.1	27	-	-	7.9	11300	8660	-	-	0.14	0.20	0.15
11-3	1243	11.9	25	3.21	1.24	5.0	9200	5780	0.52	0.20	0.08	0.15	0.094
11-4	0947	11.8	29	3.73	1.60	6.9	11500	6900	0.70	0.30	0.13	0.22	0.13
11-5	0914	11.6	29	3.63	1.36	6.2	10100	5500	0.68	0.26	0.12	0.19	0.10
11-6	0936	11.5	28	3.08	1.03	4.6	7840	3500	0.56	0.19	0.08	0.14	0.064
11-7	1021	11.5	37	2.20	0.74	3.0	5820	2200	0.53	0.18	0.07	0.14	0.053

FIG. 4-2(a)
FISSION GAS RELEASE RATES
19 SEPT - 2 OCT 1958

FIG. 4-2(b)
FISSION GAS RELEASE RATES
2 OCT - 15 OCT 1958

FIG. 4-2(c)
FISSION GAS RELEASE RATES
15 - 28 OCTOBER 1958

FIG. 4-2(d)
FISSION GAS RELEASE RATES
27 OCT - 7 NOV 1958

When reactor power was lowered, however, an additional effect was observed. Immediately after the power was lowered, all release rates fell to a very low level. Then the shorter-lived gases rose to their equilibrium levels, but the release rates of the long-lived gases rose over the next day or so to new levels which were much higher than the original levels. Then over the next few days the release rates fell off slowly to new values.

The initial decrease in release rate was first observed when reactor power was lowered from 26 to 20 Mw on October 17. It was thought that this effect might be due to a poor sample. Thus when the reactor power was lowered from 20 to 10 Mw on October 30, very great care was taken with the sampling and analysis. The initial decrease was again present and it is believed that this is a real effect.

Fission gas release rates will be discussed in more detail in Section 5.

4.5 Fission Products in Reactor Water

4.5.1 Effect of Ion Exchange Resins

A flow of about 10 gpm was maintained through the ion exchange circuit. Actually in any one experiment this rate slowly fell off, due mainly to the clogging of filters located before the resins by corrosion products.

The resins used in EBWR proved very efficient in removing dissolved ionic material from the reactor water. The decontamination factor (DF) across the resin may be defined as the ratio of the influent to the effluent specific activity.

The DF values for the resin were calculated from the water samples, which were counted for beta decay. To ensure uniformity of results, the DF was calculated always on the activities of samples two hours after sampling.

Table 4-5 shows typical results obtained during Test 1. The DF increased steadily as the test proceeded. The reason for this is not known, but spot checks taken throughout Test 2 showed a DF of $> 5 \times 10^3$ throughout the test.

The efficiency of the resins (ϵ) for removing dissolved ions may be expressed as

$$\epsilon = \frac{\text{specific activity of influent} - \text{specific activity of effluent}}{\text{specific activity of influent}}$$

Thus,

$$\epsilon = 1 - \frac{1}{DF}$$

Table 4-5

DECONTAMINATION FACTOR ACROSS
RESIN BED FOR TEST NO. 1

Date	Time	Activity of Reactor Water (c/min/ml)	Activity of Ion Exchange Effluent (c/min/ml)	DF
6-11	0942	4.5×10^5	1.7×10^3	265
6-11	1725	4.5×10^5	1.9×10^3	235
6-23	0926	5.2×10^5	5×10^2	1000
6-25	0939	5.2×10^5	6×10^2	870
6-30	1412	5.0×10^5	2.5×10^2	2000
7-7	0918	5.0×10^5	-1×10^2	5000
8-26	1038	5.2×10^5	900	600
8-28	0945	5.1×10^5	$<1 \times 10^2$	>5000
9-24	0930	5.3×10^5	$<1 \times 10^2$	>5000

From the data of Table 4-5, for all nuclides except the fission gases, $\epsilon = 1$ during these tests.

Now the rate at which a given nuclide is removed from the reactor water will be given by

$$(r\epsilon/V) N$$

where

N = Number of atoms present in reactor water

r = Flow rate to ion exchange units

V = Volume of circulating water in reactor

ϵ = Efficiency of resins in reactor = 1.

Thus for a given nuclide which is released to the reactor water at a rate R atoms/sec

$$\frac{dN}{dt} = R - \lambda N - \frac{r}{V} N$$

The magnitude of r/v is about $5.6 \times 10^{-5} \text{ sec}^{-1}$ at 10-gpm flow. (This corresponds to a "clean-up half-life" of about $3\frac{1}{2}$ hr.) This value is considerably larger than λ for all nuclides with half lives much longer than a few hours. This means that the rate of approach to steady-state activities in the reactor water will be governed, not by the rate of decay of that nuclide, but by the rate of cleanup on the resins. For a steady release rate R , all nuclides should have reached their steady-state levels after one day steady operation at a given power.

To illustrate, the above equation is easily integrated, with boundary condition $N = 0$, at $t = 0$, to give

$$N = \frac{R}{\lambda + \frac{r}{v}} \left(1 - e^{-[\lambda + (r/v)]t} \right).$$

For the case of I^{131} ($t_{1/2} = 8.05 \text{ d}$, $\lambda = 9.94 \times 10^{-7} \text{ sec}^{-1}$), if there were no ion exchange flow, I^{131} activities would build up slowly over a period of about 50 days to their saturation level, $N = R/\lambda$ atoms. For a flow of 10 gpm, a saturation level of $N = R/[\lambda + (r/v)]$ atoms would be reached in only one day.

Thus steady-state levels of activity are soon reached when the ion exchanger is in operation. The magnitude of this steady-state level will depend on the ion exchange flow rate. During these experiments this rate was changing slowly but continuously and hence, for proper intercomparison of results, all results must be adjusted for ion exchange flow rate.

In this report all rates will be corrected to zero ion exchange flow by multiplying the observed activity by the factor $[\lambda + (r/v)]/\lambda$. The activity thus obtained is the ultimate steady-state activity which would be reached if the reactor were operated for long enough steady power with no ion exchange flow.

Table 4-6 shows the results of radiochemical analyses of the reactor water for various nuclides.

The reactor had been at steady power at least 24 hr before any sample was taken. In this table the activities, corrected to zero ion exchange flow, are also shown.

Examination of this table shows three facts:

(1) All activities are extremely low in all tests. In fact, these were several orders of magnitude below the level of activation products such as Na^{24} , Mn^{56} , and Co^{58} in the reactor water. Typical steady-state

Table 4-6

STEADY-STATE FISSION PRODUCT ACTIVITIES IN REACTOR WATER AT 20 Mw

[All activities expressed as d/(min)(ml) and are corrected to time of sampling]

Date	Reactor Power (Mw)	Ion Exchange Flow Rate, gpm	Measured Activities										Activity Corrected to 0 Ion Exchange Flow											
			Cs ¹³⁸	I ¹³¹		I ¹³³		Mo ⁹⁹		Sr ⁸⁹		Ba ¹⁴⁰		Cs ¹³⁸	I ¹³¹		I ¹³³		Mo ⁹⁹		Sr ⁸⁹		Ba ¹⁴⁰	
				A	A	B	A	B	A	B	A	B	A		A	B	A	B	A	B	A	B	A	B
3-26	20	9.5		5				79		7		No Defect							1600	2500			1800	
3-31	20	9.5	148	7				89		2		16		170	380			1800	720			1800		
4-3	20	0*		50				137		5		123			50			137	5			123		
5-1	20	7	300	6								Test No. 1			240									
6-9	20	5			19			96		8		30				560			1050		1500		1700	
6-13	20	7	360	9				82	3			23	30	400	370			1206	780		1800		2400	
6-19	20	7						90		6		31						1300		1600			2500	
6-24	20	7	490	12	24			138		8		32	550	490	980			2100		2100			2600	
7-1	20	6	260	26	17			79	93	5		28	45	280	910	600		1000	1200	1100		1900	3000	
7-28	20	9	122	17	33			82	89	17		Test No. 2			900	1740			1600	1700	5800		2900	
8-7	20	9		31						6		29	140		1630								2400	
8-8	20	9	453					200				24	520					3800			2100			
8-13	20	8						70										1200						
8-14	20	8		25	22			63	144	7		33		1160	1000			1050	2400		2100		3000	
8-18	20	8		19				81						890				1300						
8-19	20	8			18				102	7		29			840				1700		2100		2600	
8-26	20	8		29	19			90	144	6		30		1350	890			1500	2400		1800		2700	
9-2	20	7		10	22			99	141	14		30		410	900			1500	2100		3700		2400	
9-8	20	7		11				77	127	14		17		450				1150	1900		3700		1350	
9-16	20	6		27				108						960				1400						
9-30	20	6	259	26				52					280	930				670						
10-8	23	6		15										540										
10-22	20	6		25				88						890				1100						
10-28	20	4.5	342	24				70					370	630				710						
10-30	20	4		14	48	323	512							330	1130	1100	1750							
(0856)																								
10-30	11	4		17	70	430	602							400	1650	1450	2050							
(1247)																								
10-31	11	3.8		14		244								310		800								
10-31	11	3.8		14		236								310		770								
11-1	11	3.5				155										480								
11-3	11	3.1			54	148	447								1100	420	1280							
11-4	11	3.0		24	13	404								430	230	1140								
11-5	11	3.0		23	36									410	640									
11-7	11	2.9		20		426								350		1160								

*Resins had been valved off 2-1/2 days previously. Therefore longer lived activities were far from saturation.

activities in d/(min)(ml) for these three nuclides at 20 Mw and at 10-gpm ion exchange flow were:

Na ²⁴	2×10^5
Mn ⁵⁶	6×10^4
Co ⁵⁸	4×10^3

(2) The results were extremely erratic. The columns labeled A and B are the results from two groups of analysts, both using the same radiochemical procedures. The agreement between results of samples is poor, as is the agreement from day to day. The most probable cause for this lack of consistency is the very low activity level.

(3) The third most noticeable effect is that in Test 1 corrected activities were not significantly higher than when no defect was present. During Test 2, activities due to I¹³¹ were in general higher; activities of Ba¹⁴⁰ and Sr⁸⁹ were slightly higher; and the activity of Mo⁹⁹ was no higher than when no defect was present.

These activities will be discussed further in Section 5.2.

4.6 Examination of Metal Specimens from Steam Line and Condenser

Gamma-ray spectra obtained from all the metal specimens from the steam line and from the turbine after each test showed the presence of only such activation products as Co⁵⁸ and Co⁶⁰. No γ -ray peaks which could be ascribed to any fission products were observed. It was concluded that any fission product activities deposited on these specimens must be very small compared with the Co⁵⁸ activity.

4.7 Examination of Fuel Specimens after Irradiation

When the fuel specimens were removed from the fuel elements, they were transferred to the Metallurgy Cave where they were cut transversely at various lengths along the specimen axis.

Typical photographs are shown in Fig. 4-3.

Examination showed that all specimens from Test 1 had cracked.

In the specimen from Test 2, pellets from the top one-foot length of the tube were uncracked. Then cracking became progressively worse until the point of maximum flux, approximately 18 in. above the bottom of the fuel, was reached. At this point, however, only severe cracking was noticeable - there were no signs of melting or recrystallization.

From this point to the bottom of the fuel cracking was progressively less and the bottom pellet was again uncracked.

Thus very roughly the extent of cracking followed the thermal neutron flux distribution and hence the temperature distribution along the length of the fuel element. Where temperatures were lowest, the fuel survived the irradiation intact and cracking appeared only in the zones of higher temperature.

FIG. 4-3-A

TEST NO. 1

VIEW OF CROSS-SECTION OF FUEL ELEMENT
AT POSITION OF DEFECT. NOTE CRACKING
BUT NO SIGNS OF RECRYSTALLIZATION.

TOP OF ROD - IN VICINITY OF DEFECT.
NOTE NO CRACKING.

40 IN. ABOVE BOTTOM OF ROD.
NOTE FIRST SIGNS OF CRACKING.

16 IN. ABOVE BOTTOM OF ROD.
APPROXIMATELY REGION OF MAXIMUM
FLUX.
NOTE SEVERE CRACKING.

BOTTOM PELLET OF ROD.
NOTE NO CRACKING.

FIG. 4-3-B
TEST NO. 2
CROSS SECTIONS OF FUEL AT VARIOUS ELEVATIONS.

5. DISCUSSION

5.1 Release Rates of Fission Products

5.1.1 No Defect - Fission Gases

When no defective fuel element is present, fission gases most probably come from fission recoils emitted from uranium oxide contamination on the surface of the fuel elements or on internal reactor surfaces in the neutron flux. The rate of release of fission recoils can be calculated from a given mass of uranium if the following assumptions are made:

(1) The uranium oxide is spread in a very thin layer of thickness less than the range of a fission fragment in the oxide, i.e., less than a thickness of $5-8 \times 10^{-4}$ cm. Even in such a film only 25% of all fission fragments formed will reach the water.

(2) Any fission gas precursor (Se, Te, Br, or I) remains in the reactor water or on a surface of the reactor and is not stripped to the steam. (During the tests conducted, no significant amounts of radio-iodines or radiobromines were detected in the reactor steam or found deposited on metal samples in the steam line or condenser.)

(3) Fission gases are stripped completely from the reactor water. In a previous report,⁽⁷⁾ this was shown to be a good assumption for the BORAX-IV reactor. That it is also a reasonable assumption for EBWR may be shown by the following calculation:

When the reactor was operated at 20 Mw, Xe^{138} was released at a rate of about 1.0 curie/day, corresponding to 6.3×10^8 atom/sec. If no Xe^{138} were stripped from the water but all decayed to Cs^{138} there, then the Cs^{138} steady concentration may be calculated to be about 3.4×10^3 d/(min)(ml) at zero ion exchange flow. The highest value found in the water was about 500 d/(min)(ml) (Table 4-6), and was generally lower. This would indicate that at least 86% of the Xe^{138} is stripped out, and the percentage may be considered to be even greater when it is considered that a certain amount of Xe^{138} is decaying in the condenser and the resultant Cs^{138} is carried back to the reactor via the condenser hotwell and feedwater.

Now, the rate of production of fission gas in the uranium oxide contamination will be

$$R' = M^{235} FY,$$

where

M^{235} = mass of U^{235} (mg)

F = fission rate/mg of U^{235} [fissions/(sec)(mg U^{235})]

Y = fission yield of nuclide in question.

It will be assumed that the release rate to the reactor water is

$$R = R'/4$$

Then

$$R = M^{235} FY/4$$

or

$$R/Y = M^{235} F/4$$

Since M^{235} and F are both constants, the ratio R/Y should be a constant value for all fission gases.

From Table 4-1, the average release rates for various fission gases may be calculated, giving the quotients R/Y shown in Table 5-1. The values of R/Y for all five gases, ranging in half-life from 17 min to 5.3 days, are constant within $\pm 15\%$.

In EBWR operating at 20 Mw a value of F is about 8.7×10^9 fissions/(sec)(mg U^{235}). Using this value, the weight of U^{235} contamination in the reactor is about 5 mg, if it is assumed that only 25% of the fission fragments reach the water.

Table 5-1

FISSION GAS RELEASE RATES;
NO DEFECT

	Release Rates at 20 Mw		R/Y
	curie/day	atom/sec	
Xe ¹³⁸	1.0	6.3×10^8	1.1×10^{10}
Kr ⁸⁸	0.053	3.35×10^8	0.93×10^{10}
Kr ^{85m}	0.012	1.17×10^8	0.90×10^{10}
Xe ¹³⁵	0.034	6.98×10^8	1.1×10^{10}
Xe ¹³³	0.0028	7.90×10^8	1.2×10^{10}
	Average: $(1.05 \pm 0.15) \times 10^{10}$		

5.1.2 No Defect - Other Fission Products

Next, it will be assumed that I^{131} and Mo^{99} are released at a rate corresponding to $R/Y = 1.0 \times 10^{10}$ fissions/sec. Neither of these nuclides will be stripped from the water, but will decay there; at steady state

$$\frac{dN}{dt} = R - \lambda N = 0$$

Table 5-2 shows the resultant steady-state activity, calculated for zero ion exchange flow.

Table 5-2

CALCULATED STEADY-STATE I^{131} AND Mo^{99} ACTIVITIES,
ASSUMING $R/Y = 1.0 \times 10^{10}$ FISSIONS/SEC

Nuclide	Y	Calculated Activity, d/(min)(ml)	Measured Activity, d/(min)(ml) (Table 4-6)
I^{131}	0.029	1500	200 to 400
Mo^{99}	0.061	3200	~1700

The measured Mo^{99} values are low by a factor of 2 when compared to the calculated value, and the I^{131} values are similarly low by a factor of 4 to 7. The reason for this discrepancy is not known, but the difference may be due to both nuclides depositing to a considerable extent on surfaces and not remaining completely in solution.

5.1.3 During Defect Tests - Fission Gases

From the data of Table 4-4 and Fig. 4-2, steady release rates of the various fission gases at steady power during Test 2 may be obtained. These values are listed in Table 5-3. Only values which were consistent over many days of operation were taken. For Xe^{135} and Xe^{133} no data for operation at 10 Mw could be obtained since the release rates fell steadily over the operating period.

In Table 5-3 the observed release rates are corrected by subtracting the release rate that was observed when no defect was present. It was assumed that the background release rate was proportional to reactor power. The release rate at 20 Mw was used to determine the proportionality constant.

Table 5-3

FISSION GAS RELEASE RATES - TEST 2

Nuclide	Xe ¹³⁸	Kr ⁸⁸	Kr ^{85m}	Xe ¹³⁵	Xe ¹³³
Fission Yield, per cent	0.057	0.036	0.013	0.064	0.065
Observed Release Rate, curie/day					
10 Mw	0.5	1.4	0.07	-	-
20 Mw	1.2	0.43	0.18	0.13	0.035
23 Mw	1.4	0.59	0.22	0.15	0.043
26 Mw	1.6	0.62	0.23	0.175	0.054
Release Rate - No Defect, curie/day					
10 Mw	0.5	0.027	0.006	0.017	0.0014
20 Mw	1.0	0.053	0.012	0.034	0.0028
23 Mw	1.15	0.061	0.014	0.039	0.0032
26 Mw	1.3	0.069	0.016	0.044	0.0042
Corrected Rate, curie/day					
10 Mw	-	0.113	0.064	-	-
20 Mw	0.2	0.377	0.168	0.096	0.0322
23 Mw	0.25	0.529	0.206	0.111	0.0398
26 Mw	0.3	0.551	0.214	0.131	0.0498
Corrected Rate x 10 ⁸ atom/sec					
10 Mw	-	7.05	6.25	-	-
20 Mw	1.26	23.5	16.4	19.7	91.0
23 Mw	1.57	32.8	20.1	22.8	112
26 Mw	1.89	34.3	20.9	26.8	140
R/Y x 10 ¹⁰ fission/sec					
10 Mw	-	2.0	4.8	-	-
20 Mw	0.22	6.5	12.6	3.1	14.0
23 Mw	0.28	9.1	15.4	3.6	17.2
26 Mw	0.33	9.5	16.1	4.2	21.5
$1/\sqrt{\lambda}$, sec ^{1/2}	38	120	151	*	812

*See text.

The values of R in Table 5-3 are obviously very small when compared to the formation rates of the fission gases in the oxide. This is illustrated in Table 5-4, in which the formation rate has been calculated using the data of Table 3-3. Obviously only an extremely small fraction (~0.01%) of any fission gas formed in the fuel was escaping through the defect.

Table 5-3 indicates that the value of R/Y is no longer constant as it was when no defect was present, and Table 5-4 indicates a

Table 5-4

COMPARISON OF FORMATION AND RELEASE RATES
OF FISSION GASES AT 20-Mw REACTOR POWER

Nuclide	Half-life	Formation Rate, atom/sec $\times 10^{-13}$	Release Rate, atom/sec $\times 10^{-9}$	Release Rate Formation Rate ($\times 10^4$)
Xe ¹³⁸	17 m	1.6	0.13	0.08
Kr ⁸⁸	2.8 h	1.0	2.4	2.4
Kr ^{88m}	4.4 h	0.36	1.6	4.5
Xe ¹³⁵	9.2 h	1.8	2.0	1.1
Xe ¹³³	5.27 d	1.8	9.0	5.0

general trend that relatively more fission gas escapes as the half-life increases. This is illustrated more quantitatively in Fig. 5-1, in which the quotient R/Y is plotted against $1/\sqrt{\lambda}$, in which λ is the decay constant for a particular nuclide. In the case of Xe¹³⁵, a corrected value of λ is used, $\lambda' = \lambda + \hat{\sigma}\phi$, where $\hat{\sigma}$, the cross section, is 3.2×10^6 barns⁽⁶⁾ and ϕ , the thermal neutron flux, is 4.6×10^{12} n/(cm²)(sec) at 20 Mw. Thus at 20 Mw, for Xe¹³⁵ it is found that $1/\sqrt{\lambda'}$ is 168 sec^{1/2}, while at 25 Mw it is 159 sec^{1/2}. The reasons for using a corrected λ are outlined in Appendix 4.

FIG. 5-1
R/Y vs. $1/\sqrt{\lambda}$
DEFECT TEST NO. 2

Figure 5-1 shows that at 20 Mw the points for the radio-xenons fall on a reasonably straight line passing through the origin. The points for radiokryptons appear to fall on a separate line of higher slope. The same behavior is seen at 26 Mw.

In Appendix 4 it is shown that for an unsheathed fuel pellet under irradiation the relationship

$$\frac{R}{Y} = 3F \sqrt{D'} \frac{1}{\sqrt{\lambda}}$$

should hold, where the symbols are those used before, F is the number of fissions/sec and D' is the special diffusion constant defined by Booth.⁽⁸⁾ Thus the linear dependence of R/Y on $1/\sqrt{\lambda}$ shown in Fig. 5-1 is not unexpected. The higher evolution rates of kryptons than for the xenons are also not unexpected, since the diffusion rates of the smaller, lighter atoms should be higher.

In Appendix 4 it is also pointed out that in a defective fuel element, as opposed to an unsheathed pellet, two diffusion processes exist: diffusion through the oxide matrix, and diffusion in the space between the fuel and the sheath. These experiments do not indicate which is the rate-controlling process, but it is likely that for the longer-lived nuclides ($t_{1/2} > 17$ min) diffusion through the oxide is rate-controlling. For shorter-lived nuclides, the fraction decaying in the gap will increase as the half-life decreases, and there will be some limiting half-life below which none escapes.

Experiments were performed in EBWR⁽⁹⁾ in which helium or argon gas was passed over $\text{ThO}_2\text{-U}^{235}\text{O}_2$ pellets similar to the ones used in these tests. The positions of these pellets and the thermal neutron fluxes to which they were subjected were similar to those involved in the present experiments. It was found that the rates of release of Xe^{138} , Kr^{88} , Kr^{85m} , Xe^{135} , and Xe^{133} again showed the same linear dependence of R/Y on $1/\sqrt{\lambda}$. Here, since the gases were continually swept out of the gap, the diffusion through the oxide lattice would be the only operative mechanism.

5.1.4 During Defect Tests - Other Fission Products

It will be shown that I^{131} and Mo^{99} are released from the fuel defect at much lower rates than are the fission gases. This will be done by first assuming that they are released by a mechanism similar to that of fission gas release. In other words, it will be assumed that they also fit on the plot of R/Y vs $1/\sqrt{\lambda}$ found to be true for the fission gases. Thus, from Fig. 5-1 we can obtain values of R/Y , from which values of R can be computed. Then it is easy to compute the saturation activity which would be present in the reactor water with zero ion exchange flow for this rate of release to the water.

Results are outlined in Table 5-5.

Table 5-5

COMPARISON OF CALCULATED AND MEASURED
RELEASE RATES OF I^{131} AND Mo^{99}

Nuclide	$1/\sqrt{\lambda}$ sec ^{1/2}	R/Y*	R, atom/sec	Calculated Steady-state Activity, d/(min)(ml)	Observed Steady-state Activity, d/(min)(ml)
Mo^{99}	585	1.03×10^{11}	6.3×10^9	3.3×10^4	700-2400
I^{131}	1000	1.76×10^{11}	5.1×10^9	2.7×10^4	400-1600

*Values chosen are those for radioxenons. If radiokrypton values were used the discrepancy would be even wider.

Thus these two nuclides are released to the water at a rate 20 to 200 times more slowly than would be expected from the fission gas release rates.

When no defect was present, the ratio of the calculated to the observed release rates was between 2 and 7, and the lower amounts of I^{131} and Mo^{99} were attributed to deposition of these nuclides on internal surfaces. Part of the discrepancy observed when a defective fuel element is present is probably due to this effect, but it is highly doubtful whether all of it is. Thus it must be concluded that nuclides such as I^{131} and Mo^{99} without a fission gas precursor are released from the defect at a significantly lower rate than are fission gases.

5.2 Transient Effects after Power Changes

5.2.1 After Power Increase

The sharp burst of activity released during reactor startup or after increases in reactor power has been observed previously when defective ceramic fuel elements had been irradiated. It is believed that this was due to the existence of a reservoir of fission gases in the space between the fuel and the sheath. Under steady conditions, fission gases were released from this reservoir at a steady rate. When power increased, however, the temperature of the fuel was increased and the pressure inside the sheath probably increased to a value above the reactor operating pressure for a short period, thus emitting a burst of fission product gases (see also Sections 5.4.1 and 5.4.2).

On startup, of course, the gap would be initially filled with water and, when this was turned to steam, there would be a corresponding pressure rise and escape of steam through the defect, carrying with it fission product activities leached from the oxide surfaces.

5.2.2 After Power Decrease

Two effects were noticed. Immediately after power was decreased, all release rates fell sharply. Then the release rates of short-lived gases returned soon to a new steady level. Release rates of longer-lived gases continued to increase for a period of a few days, after which they fell back to new low steady rates.

The initial drop can be attributed to a brief contraction of gases within the sheath following a power decrease. However, equilibrium with reactor pressure would soon be reached again and the gases should be released at a new steady rate depending on the power level.

A possible explanation for the increasing and then decreasing long-lived fission gas release rates might be that the fuel cracks during this transient. A later report⁽⁹⁾ indicates that the increased rate might be caused by escape of longer-lived gases from pores which were originally closed but were opened after the cracking.

5.3 Comparison with Other Tests

Release rates of fission gases obtained during this test may be compared with results obtained from other reactors fueled with ceramic fuel. Data (Section 5.3.2) are published on VBWR at Valecitos, California, and on PWR at Shippingport, Pennsylvania.

5.3.1 VBWR Results

Brutschy and Osborne⁽¹⁰⁾ have reported results obtained during experiments similar to those which were performed in VBWR. Their results have been obtained with UO_2 ceramic fuel sheathed in Zircaloy-2, and have been obtained with naturally occurring defects. Their data are shown in Table 5-6.

The values of R/Y when no defect is present may be compared with the data of Table 5-1. Assuming that VBWR has roughly the same thermal neutron flux as EBWR, the background uranium contamination is slightly higher in VBWR. One interesting fact is that radioiodines were more apparent than in EBWR. This is discussed more fully later.

Table 5-6

TYPICAL FISSION GAS RELEASE RATES FROM VBWR

Nuclide	Release Rate, μ curie/sec		$R/Y \times 10^{-10}$	
	No Defect	Defect	No Defect	Defect
Xe ¹³⁸	43.8	38.4	4.15	3.65
Kr ⁸⁷	4.68	6.12	4.70	6.15
Kr ⁸⁸	2.98	5.02	4.46	7.52
Kr ^{85m}	1.03	2.27	6.7	14.8
Xe ¹³⁵	1.50	11.4	4.15	31.6
Xe ¹³³	≤ 0.2	≥ 3.16	≤ 7.5	≥ 120
I ^{133(a)}	1.22	3.28	7.5	20.2
I ^{131(a)}	0.056	0.81	7.2	104

(a) Obtained from reactor water samples.

When a defect is present, a reasonable linear dependence of R/Y on $1/\sqrt{\lambda}$ is obtained, as shown in Fig. 5-2(a). However, two facts are apparent:

(1) Kryptons and xenons apparently fall on the same line, and the slope of this line is considerably steeper than that found in EBWR. In Section 5.1.2 it was shown that the slope of this line is proportional to $F\sqrt{D'}$. Values are shown below:

	EBWR (20 Mw)		VBWR,
	<u>Xenons</u>	<u>Kryptons</u>	<u>All Gases</u>
$3F\sqrt{D'}$	1.8×10^8	6.0×10^8	1.6×10^9

The reason why kryptons are released more readily than xenons in VBWR but not EBWR is not known. The higher release rate of all gases in VBWR is understandable, since the defective fuel element was operating at higher heat ratings than that in this experiment; hence D' would be greater, as well as F .

Taking an average value of F as 2.8×10^{14} fissions/sec for the fuel element in Test 2 (Table 3-3), values of D' of 4.6×10^{-14} sec⁻¹ for xenons and 5.1×10^{-13} sec⁻¹ for kryptons are obtained. Since F for VBWR is not known to the author, D' values were not calculated.

(2) Radioiodines are released to a far greater extent from the VBWR defect than from the EBWR defect. This is discussed in Section 5.4.2.

5.3.2 PWR Results

P. W. Frank⁽¹¹⁾ has analyzed the data resulting from one year of operation of the PWR reactor. The evidence indicated that one or more of the UO_2 blanket elements was defective during the latter part of the operation.

In PWR, of course, fission product gases are not stripped from solution but remain in the coolant, where eventually steady-state activities result. If fission products are released through the defect at a rate R atoms/sec and are removed from solution by radioactive decay and by cleanup on the ion exchange resins, then at steady state

$$R = \lambda N + \beta N$$

where

$$\lambda = \text{decay constant, sec}^{-1}$$

β = "clean-up" constant for ion-exchange resins.

Section 4.5.1 shows $\beta = r\epsilon/v$. For PWR a value

$\beta = 1.13 \times 10^{-4} \text{ sec}^{-1}$ was obtained from the slope of Fig. 3 (Ref. 11).

N = total number of atoms of a particular fission product in the reactor coolant.

Table 5-7 shows data from Ref. 11. Values of $R'[\text{atom}/(\text{min})(\text{ml})]$ are given rather than R because the volume of the PWR coolant is not known to the author.

Table 5-7

STEADY-STATE FISSION PRODUCT ACTIVITIES IN PWR

Nuclide	Specific Activity, d/(min)(ml)	R' , atom/(min)(ml)	$R'/Y \times 10^{-5}$	$R./Y \times 10^{-5}$ Defect Only
Cs ¹³⁹	3,000	3,270	0.50	-
Cs ¹³⁸	5,390	7,115	1.24	0.74
Kr ⁸⁷	3,870	3,370	1.55	1.05
Kr ⁸⁸	9,780	9,780	2.74	2.24
Kr ^{85m}	4,360	4,360	3.35	2.85
Xe ¹³⁵	7,670	7,670	1.20	0.70
Xe ¹³³	44,600	44,600	6.86	6.36
I ¹³³	162	22,176	3.41	2.91
I ¹³¹	1,680	18,630	6.42	5.92

In this table the value R' actually represents the sum of the released rates from the defect and from uranium contamination of surfaces in the neutron flux. In Section 5.1.1 it was shown that in EBWR, when no defect was present, R/Y was constant. It will be assumed that this is true for PWR and that no Cs¹³⁹ or Cs¹³⁸ escapes directly to the water from the fuel, but that they arise from the decay of the parent Xe¹³⁹ and Xe¹³⁸, respectively, which are released to the water. At steady state, radiochemical equilibrium will exist and activities of Xe and Cs (corrected for loss to the resins) will be equal. It will be assumed that the half-life of Xe¹³⁹ (41 sec) is short enough that virtually none will escape from a defect and that the Cs¹³⁹ present is due only to the presence of uranium contamination. Then the value of R'/Y obtained for Cs¹³⁹ should be the same for all other nuclides, for that amount of the nuclide arising from contamination and hence the value of R'/Y arising from the defect can be obtained as follows:

$$(R'/Y)_{\text{defect}} = (R'/Y)_{\text{total}} - (R'/Y)_{\text{Cs}^{139}}$$

The last column of Table 5-7 has been corrected in this manner. Fig. 5-2(b) shows the results of (R'/Y) defect plotted against $1/\sqrt{\lambda}$.

Again a reasonably linear relationship is established. Qualitatively the results are in agreement with the results of this test in that the kryptons are released at a greater rate than the xenons. However, in PWR the radio-iodines again appear much more predominantly than they did in this test.

It is perhaps significant that in this test and in the BORAX-IV experiment,⁽⁷⁾ where the fuel was a ThO_2 - UO_2 mixture, radio-iodines were not released to the coolant in any appreciable quantity, while in VBWR and PWR, where UO_2 is used, radio-iodine release rates were nearly equal to those of the fission gases. Perhaps ThO_2 retains iodine in the crystal lattice to a greater extent than does UO_2 .

5.4 Calculation of Long-lived Contamination

Although long-lived fission product contamination was not observed in the condenser or in the stack effluent, the results of this section may be used to make a rough calculation of some contamination levels. To do this, the first step is to calculate steady-state fission product gas activities in various parts of the system.

5.4.1 Steady-state Fission Product Gas Activities

It will be assumed that the reactor is operating steadily at 20 Mw. It will further be assumed that the uranium contamination in the system and the gas release rates therefrom as described in Section 5.1.1 and Table 5-1. A defective fuel rod similar to the one used in this test will be assumed present and the release rates of fission gases will be those described in the plot of Fig. 5-1. From these sets of data, the release rates of fission product gases from the background uranium and from the defect may be calculated for any fission gas. These rates, designated as R_u and R_{def} , are shown in Table 5-8.

Next, using methods fully described in Ref. 7, the steady-state activities of fission product gases in various parts of the system may be calculated. Briefly, knowing the release rate of the fission gas to the water, and assuming complete stripping from the water, the activity (A disintegrations/sec) of any nuclide may be calculated in any zone by the relation

$$A = R_{\text{in}} - R_{\text{out}} = R \left(e^{-t_{\text{in}} \lambda} - e^{-t_{\text{out}} \lambda} \right)$$

Table 5-8

EBWR FISSION PRODUCT GAS ACTIVITIES AND RELEASE RATES

Nuclide	Half-life	Release Rate to Water				Activity in Condenser, μ curie/sec		Release Rate from Stack, μ curie/sec	
		atom/sec x 10 ⁻⁸		μ curie/sec					
		R _u †	R _{def} *	from contamination	from defect	from contamination	from defect	from contamination	from defect
Kr ⁹¹	10 s	5.8	1.3	1070	249	1720	396	neg	neg
Xe ¹⁴⁰	16 s	6.4	0.54	750	64	3380	28.5	1.1	neg
Kr ⁹⁰	33 s	5.8	2.4	329	136	3920	1620	14.2	5.9
Xe ¹³⁹	41 s	6.6	0.89	301	41	6060	82	24	3.2
Kr ⁸⁹	2.6 m	4.8	4.3	58	52	1430	1290	30	27
Xe ¹³⁷	3.9 m	6.2	2.0	50	16	1290	418	32	10
Xe ¹³⁸	17 m	5.7	3.8	10.5	7.0	308	206	9.5	6.3
Kr ⁸⁷	78 m	2.5	12.3	1.0	4.9	27	133	1.0	4.8
Kr ⁸³	114 m	0.5	3.0	0.14	0.8				
Kr ⁸⁸	2.8 h	3.6	18.0	0.67	3.4	19.5	97	0.7	3.4
Kr ^{85m}	4.4 h	1.3	11.8	0.15	1.4	3.5	32	0.2	1.4
Xe ¹³⁵	9.2 h	6.4	19.7	0.36	1.1	neg	neg	0.4	1.1
Xe ¹³³	5.3 d	6.6	94.3	0.027	0.39	neg	neg	0.003	0.4

† Calculated from $R_u/Y = 1.0 \times 10^{10}$ atom/sec (Table 5-1).

* Calculated from $(R_{def}\sqrt{\lambda})/Y = 1.76 \times 10^8$ atom/sec^{3/2} for xenons and $= 6.0 \times 10^8$ atom/sec^{3/2} for kryptons (Fig. 5.1).

where t_{in} is the time a gas atom takes to get from the reactor water to the entrance to the zone and t_{out} is the time delay from the reactor water to the exit of the zone. In this calculation it is assumed that when a fission gas atom decays in a given zone its daughters remain in that zone. Values of t_{in} and t_{out} given below are taken from Table 3-2.

	t_{in} (sec)	t_{out} (sec)
Steam Dome	0	25
Piping to Turbine	25	30
Turbine to Condenser	30	60
Piping to Stack Exit	60	150

In Table 5-8 only the activities in the condenser and leaving the stack are tabulated. No data for fission product gases of half-life shorter than 3 sec are included because virtually all the gas will decay in the steam dome.

Table 5-8 illustrates several important points:

(1) The presence of one defective fuel rod such as that used in Test 2 actually released short-lived fission product gases (half-life less than 17 min) at a slower rate than they were released by the 4 mg of U^{235} contamination in the reactor. Only the longer-lived Xe^{135} and Xe^{133} were released appreciably faster.

(2) The same was true for steady-state fission product gas activities in the condenser. It would take several defects to produce as much short-lived gas activity as that present from uranium contamination.

(3) Release rates of short-lived gases from the stack, because of the delay involved, were very low, and again several defects would be necessary to approach the emission rate from contamination alone.

(4) A significant point to note is that none of the fission gases with half-life greater than that of Xe^{138} have long-lived daughter products; all are either stable or short-lived. Thus even though the longer-lived gases may be released to the condenser and from the stack in appreciable quantity, no contamination problem will result. Because of the great dilution in the outside air, the activity due to the fission product gas itself will be insignificant.

(5) Xe^{138} activities are fairly significant in the condenser. Within a few minutes after sampling, all of the shorter-lived activities will have decayed and the daughter Cs^{138} will be the predominant activity. Hence, the ubiquitous 33-minute half-life which is continually found can thus be explained.

5.4.2 N^{16} Activities

Mittl and Theys(12) give the formation rate of N^{16} ($t_{1/2} = 7.4$ sec) in EBWR as 3×10^{12} atoms/sec at 20 Mw. They show that only about 10% of the N^{16} activity is released to the steam phase. Thus, assuming that N^{16} is entering the steam at a rate of 3×10^{11} atoms/sec, it is possible to calculate the resulting N^{16} activity in the condenser by the methods of the previous section. This calculation showed the N^{16} activity in the condenser to be about 0.5 curie. Adding together all fission gas activities in Table 5-8 shows a total of only 0.02 curie of fission gas in the condenser, i.e., some 25 times less.

It is easy to see why, under steady conditions, the presence of one defective specimen made no observable change to the counting rate of the monitor at the air ejectors.

During startup or during periods when the neutron flux in the vicinity of the defective specimen was increasing, the fission gas evolution rate was increased at least by an order of magnitude during short periods, and hence their activities were now more significant and could be detected as a peak in the counting rate of the air ejector monitor.

5.4.3 Long-lived Contamination

The only decay chains which will lead to significant long-lived activities are as follows:

Using conventional methods, outlined in Ref. 9, and assuming a steady activity of fission gas in a given zone, the buildup of the daughter products may be calculated. Figure 5.3 shows the calculated buildup of long-lived activities outside the stack.

The same calculation was not done for the condenser because undoubtedly the cesiums and strontiums, which are decay products in the chain, will be washed from condenser surfaces to the hotwell and will be returned to the reactor, whence they will eventually find their way to the

FIG. 5-3
LONG LIVED CONTAMINATION FROM EBWR STACK.
BACKGROUND CONTAMINATION + 1 DEFECT.

ion-exchange resins. It was indicated in Section 4.6 that while nuclides, such as Sr^{89} , may be found in the reactor water to a very small extent, none could be detected in the steam lines or in the condenser.

The data of Fig. 5.3 were calculated on the assumption that one defective specimen and 5 mg of U^{235} contamination were present.

Obviously these amounts of activity scattered over the surrounding country will lead to negligible contamination levels. The highest contamination level after 1400 hr of steady operation, due to 51-day Sr^{89} , is roughly 7 millicuries. It must be remembered that 50% of this was a result of uranium contamination in the reactor and only the remaining 50% was due to the defect. Sr^{90} levels after the 1400-hr steady operation amounted to less than 10 microcuries, and of this only 30% was due to the defect. Thus, it is obvious that EBWR could be operated for long periods of time with only one defect such as that used in Test 2.

6. CONCLUSIONS

As a result of these tests the following conclusions may be drawn. These conclusions apply to $\text{ThO}_2\text{-UO}_2$ ceramic fuels of high density operating under the rather low heat ratings found in these tests. A small defect will be presumed present in the sheathing.

(1) The main radioactive nuclides which escape from the defect are fission product gases and the main radioactivities throughout the system are due to these gases or their decay products. The release of radio-iodines is apparently very small, much smaller, for instance, than from fuels made from UO_2 .

(2) The rate of release of gases is controlled by a diffusion-type mechanism such that the shorter-lived gases escape through the defect at a much lower rate relative to the longer-lived ones. If $R(\text{atom/sec})$ is the release rate of a fission product gas from the defect, the quotient R/Y , where Y is the fission yield, has been found proportional to $1/\sqrt{\lambda}$, where λ is the decay constant of that fission product gas. The magnitude of R is a very small fraction ($\sim 0.01\%$) of the formation rate of the nuclide.

(3) With one defect present, activities throughout the reactor system are not distinguishably different from those when no defect and only slight amounts of uranium contamination are present. In the reactor steam and even in the condenser, N^{16} activities predominate, and in the reactor water, fission product activities are orders of magnitude lower than those from activation products such as Na^{24} , Mn^{56} , or Co^{58} .

(4) If a defective element is in the close vicinity of a control rod, its presence can be detected by movement of that control rod so that the flux in the defective element is increased. If this is done, a transient increase in activity will be observed on a monitor which is measuring activities in the vicinity of the air ejector.

APPENDIX 1

THERMAL NEUTRON FLUX DETERMINATIONS

Thermal neutron fluxes for both defect tests were determined by dissolving pellets from the portions of each element which were at the point of maximum thermal neutron flux in the reactor. The solutions were then analyzed for Cs^{137} and Sr^{90} . The procedures⁽¹³⁾ detailed below were performed by S. B. Skladzien and his group.

Portions of the ceramic fuel pellets from each defect test were weighed and put into solution by refluxing in concentrated HNO_3 containing a trace of HF . These stock solutions were adjusted to an accurately known volume and, from these, aliquots were taken for Cs^{137} and Sr^{90} analyses.

The Cs^{137} was determined as the perchlorate, according to the procedure given in LA-1721.⁽¹⁴⁾ The presence of Cs^{136} in the sample was neglected, since the analysis was performed after about nine Cs^{136} half-lives had elapsed. Strontium-90, determined according to a modified procedure,⁽¹³⁾ was precipitated as the carbonate. After allowing one month for the Y^{90} daughter to grow into equilibrium, the yttrium was milked from the sample and counted. The yttrium separation procedure used was that given in UKE-CR-1003.⁽¹⁵⁾

After obtaining values for N_{Cs} and N_{Sr} , the thermal flux and total flux at 20 Mw were calculated using the following relationship:

$$\frac{N}{N_0} = Y \frac{\hat{\sigma}_{5f}}{\hat{\sigma}_5} \left(1 - e^{-\hat{\sigma}_5 \phi t} \right)$$

where:

N = Number of atoms of either Cs^{137} or Sr^{90} in sample after time t

$\hat{\sigma}_5 = \text{U}^{235}$ total cross section = 650.8 barns

$\hat{\sigma}_{5f} = \text{U}^{235}$ fission cross section = 545.0 barns

N_0 = Initial number of U^{235} atoms in sample

Y = Fission yield of either Cs^{137} or Sr^{90}

$Y_{\text{Cs}} = \text{Fission yield of } \text{Cs}^{137} = 6.1\%$

$Y_{\text{Sr}} = \text{Fission yield of } \text{Sr}^{90} = 5.9\%$

ϕ = Thermal neutron flux, $n/(\text{cm}^2)(\text{sec})$

t = Time in seconds at 20-Mw reactor

Defect Test 1 $t = 3.30 \times 10^6 \text{ sec}$

Defect Test 2 $t = 8.71 \times 10^6 \text{ sec}$

Values of $\hat{\sigma}_5$ and $\hat{\sigma}_{5f}$ were taken from Westcott.⁽⁶⁾

Results are outlined in Table A1-1. From these data the peak thermal neutron flux was taken to be $(8.0 \pm 1.0) \times 10^{12} \text{ n}/(\text{cm}^2)(\text{sec})$.

Table A1-1

RADIOCHEMICAL DETERMINATION OF
THERMAL NEUTRON FLUX

	Test 1	Test 2
N_{Cs} (atoms)	9.06×10^{13}	2.00×10^{13}
N_{Sr} (atoms)	1.04×10^{13}	2.45×10^{13}
N_0 (atoms)	1.08×10^{16}	1.10×10^{16}
$\phi t (\text{n}/\text{cm}^2)$		
from Cs^{137}	2.76×10^{19}	5.75×10^{19}
from Sr^{90}	2.99×10^{19}	7.18×10^{19}
$\phi [\text{n}/(\text{cm}^2)(\text{sec})]$		
from Cs^{137}	8.14×10^{12}	6.60×10^{12}
from Sr^{90}	8.82×10^{12}	8.24×10^{12}

Since the specimen used in Test 1 was small, it was assumed that the flux was uniform along its length. For Test 2, with the 4-foot length of fuel, the ratio of maximum-to-average thermal neutron flux was taken to be 1.72.*

*This value was obtained from a measurement made of the vertical thermal neutron flux distribution measured in an EBWR channel using Cs foils.

APPENDIX 2

CALCULATION OF FUEL TEMPERATURES

This appendix describes the methods used to estimate the central temperatures of the test specimens while under irradiation. The final temperatures must be regarded as quite approximate, even though a fairly involved procedure was used in their calculation. This is because no calculation can give better accuracy than that of the parameters used.

Temperatures were calculated as follows:

(1) The bulk water temperature in EBWR (T_w) was taken as 490°F (254°C). It was assumed that the surface temperature of the stainless steel sheath was 257°C. The temperature rise across the sheath was calculated using a thermal conductivity of 0.185 watt/(cm²)(°C/cm).⁽¹⁶⁾

(2) An approximate figure for the gap between the fuel and the sheath was assumed. The temperature rise across the gap was calculated assuming the gap was filled with steam. A value of the thermal conductivity at approximately the right temperature was taken from Ref. 16. Thus an approximate value for the temperature of the pellet surface (T_s) was obtained.

(3) The values of the thermal conductivity of ThO₂ were taken from Taraba.⁽¹⁷⁾ Although the fuel was a ThO₂-UO₂ mixture, the UO₂ content was low and it was assumed the thermal conductivity was that of ThO₂. From Taraba's data, a plot of

$$\int_{T_s}^{2500} k(T)dT \text{ vs } T_s$$

was drawn, as shown in Fig. A2-1. From the surface heat flux (Table 3-3), the value of

$$\int_{T_s}^{T_c} k(T)dT$$

was calculated from the relationship

$$\int_{T_s}^{T_c} k(t)dT = \frac{1}{2} Wa \quad ,$$

where W is the heat flux at the surface of the fuel (watts/cm²) and a is the radius of the fuel (cm). Then, since

$$\int_{T_c}^{2500} k(T) dT = \int_{T_s}^{2500} k(T) dT - \int_{T_s}^{T_c} k(T) dT, \quad ,$$

the value of T_c could be easily read from Fig. A2-1.

FIG. A2-1
PLOT OF $\int_{T_s}^{2500} k dT$ vs T_s FOR ThO_2

(4) Using these values of T_c and T_s , an average temperature for the fuel was calculated, and knowing the original pellet diameter, the expansion at this mean temperature was calculated using a coefficient of expansion for the ThO_2 of $1.2 \times 10^{-5} \text{ } ^\circ\text{C}^{-1}$. From these values a better value of the gap between fuel and sheath was recalculated.

(5) Using this new gap width, and with a better idea of the value of k_{steam} , the temperature rise across the gap, and the values of T_s and T_c were recalculated as in items (2) and (3) above. Then with these new sets of values, the gap width was recalculated as in item (4) and the whole process was repeated. This process of successive approximations was carried on until a consistent set of results for gap width, T_s , and T_c was obtained. Table A2-1 illustrates the results obtained for three reactor powers.

Table A2-1

FUEL PELLET TEMPERATURES DURING IRRADIATION

	Test No. 1			Test No. 2					
	10 Mw	20 Mw	30 Mw	Maximum			Average		
				10 Mw	20 Mw	30 Mw	10 Mw	20 Mw	30 Mw
W (fuel surface) (watt/cm ²)	26	52	78	24	48	72	14	28	42
$\int_{T_c}^{T_s} k(T)dT$ (watt/cm)	5	10	15	4.8	9.6	14.4	2.8	5.6	8.4
$k_{\text{steam}} \times 10^4$ [watt/(cm ²)(°C/cm)]	4.7	5.2	5.5	5.5	6.1	6.2	4.8	5.2	5.7
Gap at temperature (cm)	0.0046	0.0038	0.0028	0.0107	0.0089	0.0071	0.0114	0.0102	0.0089
T _{ss} (°C)	257	257	257	257	257	257	257	257	257
ΔT_{ss} (°C)	8	17	25	6	11	17	6	11	17
ΔT_{gap} (°C)	256	382	394	495	706	822	331	550	656
T _s (°C)	521	656	676	758	974	1096	594	818	930
T _c (°C)	610	930	1110	900	1290	1630	720	1000	1210

Because of the rather low surface heat fluxes during these tests, the temperature rise across the gap between the fuel and the sheath is an appreciable fraction of the total ΔT throughout the specimen. In Test 1, with the smaller gap, ΔT_{gap} is comparable to ΔT_{fuel} , while during Test 2 with the larger gap, ΔT_{gap} is much larger than ΔT_{fuel} . This tends to decrease the uncertainty of core temperatures to some extent in that it is believed that the thermal conductivity of the steam is better known than that of the oxide.

It is very hard to assess the limits of error in these calculations, but it is felt that the calculated central temperatures are accurate to $\pm 10\%$.

APPENDIX 3

ANALYSES OF AIR EJECTOR SAMPLES FOR FISSION PRODUCT GASES

Two methods of gas sampling and analysis were used. The first, used exclusively until the middle of September, 1958, consisted of determining the activities of Xe^{138} and Kr^{88} by allowing their daughter products Cs^{138} and Rb^{88} to grow into equilibrium with the gas sample, and then β -counting the daughters. Full details of this method, which will hereafter be referred to as the Chemical Method are found in Section 1 of this appendix.

By mid-September, 1959, a 200-channel γ -ray pulse height analyzer had been obtained and calibrated. From this time on, this instrument was used for determination of fission product gases. Full details of this method, referred to hereafter as γ -ray Pulse Height Analysis (PHA), will be found in Section 2 of this appendix.

1. Chemical Methods for Xe^{138} and Kr^{88}

1.1 Decay Schemes

The decay schemes for Xe^{138} and Kr^{88} are outlined below.

In Figs. A3-1 and A3-2 curves are plotted showing the decay of the parent activity and buildup of daughter activity. These curves were calculated on the assumption that initially the gas sample consists of pure parent (Xe or Kr) activity and that the daughter grows in from sampling time.

Decay constants used for calculation of these curves are shown in Table A3-1.

FIG. A3-1
BUILD UP AND DECAY CURVES FOR
 $\text{Xe}^{138} \rightarrow \text{Cs}^{138} \rightarrow \text{Ba}^{138}$

FIG. A3-2
BUILD UP AND DECAY CURVES FOR
 $\text{Kr}^{88} \rightarrow \text{Rb}^{88} \rightarrow \text{Sr}^{88}$

Table A3-1

DECAY CONSTANTS OF FISSION GASES
AND DAUGHTERS

Nuclide	Half Life, $t_{1/2}$	Decay Constant $\lambda (\text{min}^{-1})$
Xe ¹³⁸	17 min	0.0408
Cs ¹³⁸	33 min	0.0210
Kr ⁸⁸	2.78 hr	0.00416
Rb ⁸⁸	18 min	0.0385

The method of calculation is quite standard⁽¹⁸⁾ and will be merely outlined here. In the decay scheme

if N_i represents the number of atoms of nuclide i present in the sample at time t , then

$$\frac{dN_a}{dt} = -\lambda_a N_a \quad (1)$$

and

$$\frac{dN_b}{dt} = \lambda_a N_a - \lambda_b N_b \quad (2)$$

The solutions to these equations with the boundary condition that at $t = 0$

$$N_a = N_a^0$$

$$N_b = 0$$

are

$$\lambda_a N_a = \lambda_a N_a^0 e^{-\lambda_a t} \quad (3)$$

and

$$\lambda_b N_b = \lambda_a N_a^0 \frac{\lambda_b}{\lambda_b - \lambda_a} (e^{-\lambda_a t} - e^{-\lambda_b t}) ,$$

where $\lambda_a N_a$ and $\lambda_b N_b$ are, of course, the activities of a and b expressed in disintegrations per unit time.

The curves in Figs. A3-1 and A3-2 are plots of the ratios $\lambda_a N_a / \lambda_a N_a^0$ and $\lambda_b N_b / \lambda_a N_a^0$.

1.2 Sampling Method

Several methods of sampling were tried before a final procedure was decided upon. Only this procedure will be described here.

The sampling apparatus on the air ejector gas piping is shown schematically in Fig. A3-3. A small piece of polythene tubing was attached permanently to the outlet of the sampling valve. A wad of glass wool in the polythene tube was used to ensure that no Cs^{138} or Rb^{88} which might be carried by the gas stream entered the sampling tubes.

FIG. A3-3
SAMPLING METHOD FOR Xe^{138} AND Kr^{88} SAMPLES

The two glass bulbs A and B, each about 8 in. long and 0.5 in. ID, were shaped as shown. Bulb A was used for Xe^{138} determination and Bulb B for Kr^{88} . Originally, stopcocks were used at each end of the sampling bulb; however, it was found that extreme cleanliness of the bulbs was essential, and that stopcock grease tended to trap activity. Consequently, the stopcocks were replaced by the polythene tube and pinch clamps as shown. After each determination the polythene tube was removed and the glass bulb was immersed in chromic-sulfuric acid cleaning solution until needed again. The volume of the bulb, about 100 cc, was carefully calibrated. The pinch clamps could not be placed in exactly the same position each time but it was found that the error introduced was less than +1%, which was negligible.

To take a sample, the two bulbs were joined together as shown, and were then evacuated to a pressure of ~ 1 mm Hg. Pinch clamps 1 and 4 were closed. Then the sampling train was attached to the polythene tube on the air ejector gas sampling valve as shown and the valve was opened. Pinch clamps 1 and 4 were also opened, and gas was allowed to flow through the two bulbs for about 3 min. This was found to be sufficient time to obtain a representative sample of the air ejector gases.

To avoid activity release into the atmosphere, the effluent gases from Bulb B were led to an active charcoal trap.

At the end of 3 min, pinch clamps 1, 2, 3, and 4 were tightly closed in the order 4, 3, 2, 1. The sampling valve was also closed. The train was removed and the two bulbs were separated by cutting the polythene tube between pinch clamps 2 and 3. The pressure in the air ejector gas stream at the sampling point was between 8 and 14 in. of water above atmospheric; hence, the pressure in the bulb was taken to be atmospheric.

Before and after sampling, the flow rate of the air ejector gas stream was obtained by reading of the flowmeter located in the line upstream of the sampling valve (see Section 1.5.2(2) of this appendix). The time at which pinch clamp 4 was closed was taken as the time of sampling and will be referred to as T_s .

1.3 Analytical Methods

1.3.1 Xe¹³⁸

The procedure used for Xe¹³⁸ determination will now be described. The letters in parentheses refer to notes which will be found at the end of the description.

Bulb A was allowed to stand for 30 to 45 min (a) after sampling. Then at a time noted as T_p , pinch clamps 1 and 2 were opened and the gas was purged from the bomb by a slow stream of air (b). This and all subsequent steps were carried out in a well-ventilated fume hood. Pinch clamp 2 was next closed and about 5 ml of 4N HNO₃ were run in through the top piece of polythene tube (c). Pinch clamp 1 was closed and the bulb was thoroughly shaken for a short time. This solution was then run into a 25-ml volumetric flask (d). This procedure was repeated two or three times with small additions of HNO₃, each addition being shaken and then run into the volumetric flask. The extent to which the activity was removed could be followed by monitoring the sides of the bulb with a Geiger-Muller probe. When the remaining activity in the bulb was down to a negligible level, the bulb was rinsed once or twice with deionized water, the rinsings being added to the volumetric flask (e).

The flask was then filled to the mark with deionized water and shaken thoroughly to ensure proper mixing. Two aliquots of this solution, usually 500 λ (0.500 ml), were evaporated to dryness on stainless steel counting planchets under an infra-red heat lamp and then were counted on a gas-flow proportional counter of known geometry (f)(g).

The decay of the samples was followed for sufficient time to ensure that a 33-minute half-life was present. Quite often it was found that some Rb⁸⁸ (18-minute half-life) was present. In such cases the decay was followed for sufficient time to establish the 33-minute half-life.

NOTES

(a) Figure A3-1 shows that Cs¹³⁸ activity will be at its maximum 34 min after sampling.

(b) Several tests were carried out in which the gas was purged through a glass wool filter to see whether any Cs¹³⁸ activity was removed with the gas stream. Negligible amounts were found present on the filter.

(c) It is preferable that the 4N HNO₃ contain a few mg/ml of Cs⁺ tracer. Tests showed that this was not absolutely necessary but is, nevertheless, good practice.

(d) A glass tube, with one end drawn down, attached to the polythene tube makes transfer of the acid from the bulb to the volumetric flask much easier.

(e) Unless the glass surfaces are extremely clean it will be found difficult to remove the last traces of activity. Warming the bulb during the shaking process was found to be helpful in this respect.

(f) Aliquots of standard, carrier-free solutions of several nuclides of varying β energy were evaporated to dryness on stainless steel counting trays, similar to those used for Cs¹³⁸ analysis, to calibrate and obtain the geometry (efficiency) of the counter. It was found that the efficiency was independent of β energy over the range covering Cs¹³⁸ and Rb⁸⁸ β rays. Since virtually no deposit was left after evaporation of the solution on the counting tray, no self-absorption corrections were necessary. The counter was standardized daily with a standard Co⁶⁰ source of known disintegration rate.

(g) Since the nuclide counted, Cs¹³⁸, has a half-life of 33 min, the procedure outlined above should be carried out as quickly as possible.

1.3.2 Kr^{88}

The second bulb was allowed to stand for about 4 hr before purging the gas. Figure A3-2 shows that the Rb^{88} activity will have passed its maximum value by this time, but the increased time is necessary to allow any Cs^{138} to decay to a negligible level.

After purging of the gas, the bulb was treated in a precisely similar manner to that described in the preceding section. This time, because of the short Rb^{88} half-life (18-minute) it was most important that the procedure be carried out as rapidly as possible. In nearly every case it was found that the final activity counted was pure Rb^{88} and decayed with an 18-minute half-life; there was very little evidence of a 33-minute "tail" to the decay curve.

1.4 Calculation of Specific Activity in Bulb

Figure A3-4 shows typical decay curves obtained for Cs^{138} and Rb^{88} . For both gases the calculations were done in a similar manner.

FIG. A3-4
TYPICAL Cs^{138} AND Rb^{88} DECAY CURVES
SAMPLED 1024 hrs 11-7-58

First, the decay curve was extrapolated back to the time at which the bulb was purged (T_p). For Rb^{88} this merely involved extrapolation of the 18-minute decay curve but, as mentioned previously, the Cs^{138}

decay usually had to be followed long enough for a 33-minute decay to become established. This 33-minute decay line was extrapolated back to T_p .

Let the observed counts/min of Cs^{138} at T_p be B and let the efficiency of the counter be ϵ , the volume of solution evaporated be v_a ml, and the volume of the volumetric flask be v_t ml. Then at T_p the number of disintegrations/min in the volumetric flask, and hence in the sampling bulb, will be

$$B v_t / \epsilon v_a \text{ disintegrations/min.}$$

If the volume of the bulb is V ml, the specific activity of Cs^{138} in the bulb at T_p will be

$$B v_t / \epsilon v_a V \text{ disintegrations/(min)(ml)}$$

Next, to determine the Xe^{138} activity at sampling time, Fig. A3-1 is referred to. Looking at the growth and decay curve for Xe^{138} , the value of

$$\text{Activity } Cs^{138} / \text{Initial Activity } Xe^{138}$$

is determined for the time T_p . For instance, at 35 min after sampling the Cs^{138} activity will be 0.254 of the initial Xe^{138} activity. If this value is f at time T_p , then

$$Xe^{138} \text{ activity in air ejector gases} = \frac{B v_t}{\epsilon v_a V} \times \frac{1}{f} \text{ d/(min)(ml)}$$

The activity of Kr^{88} is calculated in the same manner, except that the growth and decay curve for Rb^{88} in Fig. A3-2 is used.

The calculation of release rates from the defect is outlined in Section 3.5.1 of the text.

1.5 Assessment of Errors

1.5.1 Analytical Procedure

Analyses of air ejector gas samples taken at the same reactor power and the same air ejector gas flow rate usually agreed within about +10%. Data taken from Tables 4-2 and 4-3 and shown in Table A3-2 illustrate this.

Table A3-2

COMPARISON OF AIR EJECTOR GAS SAMPLES FOR
VARIOUS REACTOR POWERS AND GAS FLOW RATES

Date	Time	Reactor Power (Mw)	Gas Flow Rate (ℓ /min)	Activity [d/(min)(ℓ)]	
				Xe ¹³⁸ ($\times 10^{-7}$)	Kr ⁸⁸ ($\times 10^{-6}$)
6-11-58	1455	21.2	97	2.6	
6-11-58	1457	21.2	97	2.7	
6-12-58	1033	20.0	97	1.9	1.4
6-12-58	1400	20.9	97	2.4	1.8
6-19-58	1103	20.9	61	2.6	2.3
6-19-58	1348	21.4	61	2.2	2.0
6-26-58	1320	20.3	32	2.7	2.3
6-26-58	1345	20.6	32	2.2	2.3
6-27-58	0837	20.6	77	1.6	1.0
6-27-58	0911	20.6	77	1.5	0.75
6-27-58	1237	20.4	77	1.4	0.65
6-27-58	1237	20.4	77	1.7	0.90
7-28-58	0907	20.0	48	4.1	
7-28-58	1346	20.0	47	3.6	

1.5.2 Other Errors

There are several other sources of error inherent in this procedure:

(1) While sampling, as the active gas is flushing through the bulb, some Cs¹³⁸ resulting from decay of Xe¹³⁸ may settle out on the glass surface. It is believed that this will not introduce a large error since the time of flushing (2 to 3 min) was short compared to the half-life of the gas and because most decay products will probably be carried along in the gas stream.

(2) The final calculations of release rates of the fission product gases involve the flow rate of the air ejector gas stream (see Section 3.5.1 of text). The manufacturer's calibration curve was used to convert the flow meter reading to liters/min. Since this instrument was calibrated for air flow and because the air ejector gases contained varying amounts of H₂ and

O₂ mixed with the air, an analysis of the gases for H₂, O₂, and N₂ was always performed when a sample was taken. Using this analysis, the density of the gas was calculated and the necessary correction applied, again using the manufacturer's data.

Flow rate readings on this instrument were sometimes rather erratic, and this caused the largest uncertainty in the final calculation of fission gas release rates. Table 4-3 shows that in the period from August 11 to September 15, 1958, when reactor power was constant at 20 ± 0.6 Mw, fission gas release rates agreed within $\pm 20\%$ for both Xe¹³⁸ and Kr⁸⁸.

2. Gamma-ray Pulse Height Analyses

2.1 General

The equipment used for obtaining the gamma-radiation information presented in this report is as follows: A 3 in. by 3 in. right cylinder sodium iodide scintillation detector on a 3-inch DuMont phototube, and a conventional preamplifier were located in an iron-shielded counting cave with inside dimensions of 2 ft wide, 3 ft long, and $2\frac{1}{2}$ ft high. The iron thickness was 6 in.

A 200-channel analyzer,* associated linear amplifier, recorder, and IBM typewriter readout were used for pulse analyses. Information accumulation was made using a Live Timer which automatically corrected the sample analysis time for instrument dead-times.

2.2 Sampling Method

Several glass bulbs with two-way stopcocks, as shown in Fig. A3-5, were prepared. Care was taken that each bulb was as similar in dimensions, thickness of glass, etc., as possible. The volume of each bulb was calibrated by weighing with the bulb completely full of distilled water at a known temperature. All bulbs held the same volume within ± 0.5 ml.

Before sampling, the bulb was evacuated to a pressure of about 1 micron of Hg and the stopcock was closed. The bulb was then attached to the polythene tube on the air ejector sampling valve, as shown in Fig. A3-5. The glass wool plug was always kept in this tube to filter out Cs¹³⁸ and Rb⁸⁸.

The stopcock was then positioned for flushing, as shown in Fig. A3-5, the air ejector sample valve was opened. The gases were again led to the active charcoal trap to prevent contamination of the atmosphere.

*Radiation Instrument Development Laboratory, Chicago, Illinois, Model 3302.

FIG. A3-5
SAMPLING METHOD FOR AIR EJECTOR
GAS SAMPLES

When the tube and the inside of the stopcock had been thoroughly flushed (1 or 2 min) the stopcock was turned so that there was direct access to the bulb from the sampling valve. It was left in this position for a few seconds for pressure to equilibrate and then was closed. The sampling valve was also closed. The time at which the stopcock was closed was noted as the sampling time T_s . The pressure of the gas in the air ejector line and the flow rate of this line were recorded before and after sampling. If they were not constant over

this period, another sample was taken. The pressure was never more than 10 to 12 in. of water above atmospheric and hence the pressure of the gas in the bulb was taken to be 1 atmosphere.

The bulb was then removed from the sampling valve and the upper tube, and the inside of the stopcock, and the outside of the bulb were thoroughly washed to remove any contamination. The bulb was wrapped in one layer of Saran Wrap and was taken to the gamma-ray pulse height analyzer.

2.3 Gamma-Ray Pulse Height Analysis

2.3.1 Sample Counting

A special holder was constructed so that the sampling bomb could be placed on top of the 3 in. by 3 in. scintillator in a reproducible position. Gamma-ray spectra were run at various times after sampling at the instrument settings shown in Table A3-3.

Table A3-3

DATA FOR NUCLIDE DETERMINATION BY PULSE HEIGHT ANALYSIS

Count No.	Time after Sampling	Energy Range Covered (Mev)	kev/channel	Nuclides Determined
1	35 min	0.2-2.2	10	Cs^{138} , Kr^{88}
2	4 hr	0.7-2.7	10	Kr^{88}
3	20 hr	0-0.5	5	Xe^{133} , Xe^{135} , Kr^{85m}
4	2 days	0-0.5	5	Xe^{133}

Typical spectra are shown in Figs. A3-6, A3-7, and A3-8.

In each case spectra were counted for a sufficiently long time that the best statistics were obtained without the counting time being too long in comparison with the half-life of the nuclide to be determined. For instance, during Count No. 1, 10 minutes for counting was usually sufficient while, for Count No. 2 a counting time of at least 20 minutes was usually necessary. For Counts Nos. 3 and 4 counting times of one-half hour were usually sufficient.

2.3.2 Analysis of Spectra

In all cases, the counting rate for each fission gas was determined by summing the area under a given γ -ray peak by a purely arbitrary method. For a given nuclide the same method was used throughout the test. Several standardizations were made in which the counting rate of a given nuclide in a given sample was determined, and at the same time the absolute disintegration rate of that same nuclide, in a sample taken at

FIG. A3-6
 γ -RAY SPECTRUM OF GAS SAMPLE
 30 MINUTES AFTER SAMPLING

FIG. A3-7
 γ-RAY SPECTRA OF GAS SAMPLES
 4 HOURS AFTER SAMPLING

the same time as the first, was measured by another method. Details will be found in Section 2.4 of this appendix. Thus, a factor to convert from counting rate to disintegration rate was obtained. Activities of nuclides were also corrected to sampling time. Methods for the determination of the various nuclides are given below.

(a) Kr⁸⁸

The 2.4-Mev γ -ray peak was observed on spectra obtained 4 hr after sampling. The total counts under the peak were determined as follows:

FIG. A3-8
 γ-RAY SPECTRUM OF GAS SAMPLE
 20 HOURS AFTER SAMPLING

The peak maximum was first determined. For high counting rates this presented no difficulty, but for lower counting rates and poorer statistics this was done by estimating the midpoint of the peak. Let this be Channel A. Then the counts in all channels from A to the channel with the minimum count, B, were summed. The background (which was counts in Channel B x number of channels summed) was then subtracted and the results multiplied by 2. In other words,

$$\text{counts } (C_K) = 2 \left[\sum_A^B \text{counts in channel} - (B - A + 1)(\text{counts in Channel B}) \right]$$

From this the count rate could be determined by count rate (CR) = counts/time counted.

The disintegration rate at the sampling time was then calculated from

$$\text{Disintegration rate [d/(min)(ml)]} = CR f_K e^{\lambda_{Kr} \tau} / V,$$

where

f_K = Factor for converting count rate to disintegration rate (Section 2.4).

λ_{Kr} = Decay constant for Kr^{88} , sec^{-1} .

τ = Time elapsed between sampling and counting, sec.

V = Volume of sample counted, ml.

The factor $e^{\lambda_{Kr} \tau}$ was found from Fig. A3-2 using the curve for Kr^{88} decay to find $e^{-\lambda_{Kr} \tau}$ and then taking the reciprocal.

(b) Xe^{138}

The spectra taken 30 min and 4 hr after sampling were used. Figure A3-6 shows that the Cs^{138} peak at 1.43 Mev is contaminated with Kr^{88} , and hence a correction factor must be applied. This was done as follows.

First, the number of counts under the entire peak was determined.

$$\text{Total counts } (C_T) = \sum_A^B \text{ counts in channel} - (B - A + 1)(\text{counts in Channel B}).$$

Next, it was determined from a large number of spectra that the ratio of the counts under the 1.55-Mev peak to the counts

under the 2.4-Mev peak for Kr^{88} was 0.6 ± 0.1 . Counts under the peak were determined as in Section 2.3.2(a) above.

Thus the counts under the 2.4-Mev peak at time τ_2 when Count No. 2 was taken (usually 4 hr after sampling) were used to find the correct number of counts which would be present under the 1.55-Mev peak at time τ_1 when Count No. 1 was taken (usually 30 min after sampling):

$$C_{K(1.55)} = 0.6 C_{K(2.4)} e^{\lambda(\tau_2 - \tau_1)} \times \frac{\text{time counted for Count No. 1}}{\text{time counted for Count No. 2}} .$$

Then the number of Cs^{138} counts under the 1.43-Mev peak were obtained:

$$C_{Cs} = C_T - C_{K(1.58)} ,$$

and the count rate:

$$CR_{Cs} = C_{Cs} / \text{time counted} .$$

The disintegration rate of Xe^{138} at sampling time was then calculated by the equation

$$\text{Disintegration rate [d/(min)(ml)]} = CR_{Cs} f_{Cs} / V R ,$$

where f_{Cs} and V have the same significance as before and R is the ratio $\lambda_{Cs} N_{Cs} / \lambda_{Xe} N_{Xe}^0$ determined at time τ_1 from Fig. A3-1.

(c) Xe^{135} , Xe^{133} , and Kr^{85m}

The spectra taken 20 hr and 2 days after sampling were used. The activity of the isotope Xe^{135} was determined from the 250-kev, Xe^{133} from the 81-kev, and Kr^{85m} from 150-kev γ -ray peak. In all instances counting rates were high enough and the peaks were symmetrical enough that the count under the peak could be determined by the equation

$$\text{Counts} = \sum_A^B \text{ counts in channel} - (B - A + 1)(\text{counts in Channel B}) \text{ as before.}$$

Then the count rate, (CR) = counts/time counted, and disintegration rate at sampling time are connected by the relation

$$\text{disintegration rate} = \frac{(\text{CR}) f e^{\lambda \tau}}{V} \text{ d/(min)(ml)} .$$

Disintegration rates at sampling time having been obtained, the release rates of the various fission gases from the defect were calculated as in Section 3.5.1 of the main report.

2.4 Standardization

2.4.1 Xe¹³⁸ and Kr⁸⁸

The spectrometer was standardized for Xe¹³⁸ and Kr⁸⁸ as follows:

Simultaneous samples of the air ejector gas stream were taken for chemical and for γ -ray spectrum analyses by the methods of Sections 1.2 to 1.4 and of Sections 2.1 to 2.3 of this appendix, respectively. Thus the absolute disintegration rate obtained by the first method could be compared to the counting rate obtained by the second. Typical results are given in Table A3-4.

2.4.2 Xe¹³⁵ and Xe¹³³

Standardization for Xe¹³⁵ and Xe¹³³ was carried out by taking simultaneous samples by the methods described in Section 2.2 of this appendix. One sample was counted on the γ -ray spectrometer. Carrier xenon was added to the other and the xenon fraction was separated by a gas chromatographic technique, and was counted in a gas-counting apparatus of known geometry and efficiency. The resulting decay curve was resolved into two components, that due to Xe¹³⁵ ($t_{1/2} = 9.2$ hr) and that due to Xe¹³³ ($t_{1/2} = 5.27$ days). By extrapolating back to sampling time, the disintegration rates for the two nuclides could be obtained. Accuracy of this gas-counting method is reported to be better than $\pm 5\%$. The results of these determinations are listed in Table A3-5.

Table A3-4

A COMPARISON OF THE CHEMICAL AND GAMMA-RAY
SPECTRUM METHODS FOR Xe^{138} AND Kr^{88} ANALYSIS

Chemical Methods, d/(min)(ml)	γ Spectrum, counts/(min)(ml)	Ratio, disintegrations/ count
Xe^{138}		
7.8×10^5	2.0×10^4	38
2.9×10^6	4.7×10^4	62
3.1×10^6	5.5×10^4	56
8.7×10^5	1.8×10^4	48
2.7×10^6	4.4×10^4	61
2.8×10^6	7.5×10^4	37
7.9×10^5	2.1×10^4	38
		Average 49 ± 12
Kr^{88}		
2.4×10^6	2.1×10^4	114
3.5×10^6	2.9×10^4	121
2.0×10^5	1.7×10^3	118
5.6×10^5	4.3×10^3	130
3.5×10^5	2.6×10^3	135
1.6×10^5	1.5×10^3	107
4.0×10^5	3.3×10^3	121
1.5×10^6	1.4×10^4	107
		Average 120 ± 15

Table A3-5

A COMPARISON OF THE GAS COUNTING AND
GAMMA-SPECTRUM METHODS FOR
 Xe^{133} AND Xe^{135} ANALYSIS

Gas Counting, d/(min)(ml)	counts/ (min)(ml)	disintegrations/ count
Xe^{133}		
7.3×10^4	5.3×10^3	13.8
5.1×10^4	3.3×10^3	15.5
9.7×10^4	6.6×10^3	14.7
		Average 14.7 ± 0.9
Xe^{135}		
2.1×10^5	3.7×10^4	5.7
1.2×10^5	1.7×10^4	7.0
2.7×10^5	4.0×10^4	6.8
1.5×10^5	2.2×10^4	6.8
		Average 6.6 ± 0.6

2.4.3 Crystal Efficiency - $\text{Kr}^{85\text{m}}$ Standardization

An indirect method was used to determine the disintegration rate from $\text{Kr}^{85\text{m}}$ counting rates. From the data of Section 2.4.1 above, a measure of the crystal efficiencies for the various γ -ray energies noted below were calculated. Data obtained for the 4 in. by 4 in. crystal are outlined in Table A3-6. The values of N , α , and Ng are taken from Seaborg's Tables.⁽¹⁹⁾

Table A3-6

PARAMETERS FOR CRYSTAL EFFICIENCY DETERMINATION

Nuclide	γ -Ray Energy (Mev)	Ratio (d/counts)	N	α	Ng	ϵ
Kr^{88}	2.40	120	0.35	-	0.35	0.024
$\text{Xe}^{138} (\text{Cs}^{138})$	1.43	49	0.73	-	0.73	0.028
Xe^{135}	0.25	6.6	0.97	0.054	0.92	0.165
Xe^{133}	0.081	14.7	1.00	1.5	0.40	0.170

In Table A3-6,

N = Number of γ rays of the given energy emitted from the nucleus per disintegration

α = Internal conversion coefficient (e/γ in Seaborg's nomenclature)

Ng = Number of γ rays of the given energy emitted from the atom per disintegration.

$$\text{Ng} = N \frac{1}{1 + \alpha}$$

ϵ = "Efficiency" of the crystal - defined as the ratio of the number of counts registered by the counter to the number of γ rays of a given energy emitted by the sample:

$$\epsilon = 1/\text{NgR}$$

It will be noted that the value of ϵ is constant, within experimental error, in the energy range from 0.08 to 0.25 Mev. The 0.15-Mev γ ray of $\text{Kr}^{85\text{m}}$ falls in this range. For this nuclide,

$$N = 0.77$$

$$\alpha = 0.040$$

$$\text{Ng} = 0.74$$

Hence, taking

$$\epsilon = 0.17,$$

it was found that R , disintegrations/counts = 8.0.

Thus the $\text{Kr}^{85\text{m}}$ disintegration rate was determined from the count rate under the 0.15-Mev γ -ray peak for use of this factor.

2.5 Assessment of Errors

Even though the method of analysis was different, the final calculations of release rates of fission gases from the defect still involved the flow rates of the air ejector gases. Since these were erratic, an accuracy of no better than $\pm 20\%$ can be claimed for the observed release rates.

Actually, reproducibility was rather better than this. Table 4-4 shows that in the period from September 27 to October 6 when the reactor power was steady and steady conditions prevailed, release rates of all fission gases were reproducible to better than $\pm 15\%$.

If a better means of measuring air ejector flows were adopted, it is felt that results should be accurate to better than $\pm 10\%$ and reproducible to an even lower figure.

APPENDIX 4

DIFFUSION OF FISSION GASES FROM CERAMIC FUEL

1. General Treatment

The model used for these calculations will be that proposed by Booth.^(8,20,21) The mathematical procedure is identical with that proposed by Litz.⁽²²⁾

The diffusion model is taken as a sphere of radius a cm in which the fission gas is being generated at a constant rate B' atoms/(sec)(cm³). Back diffusion of the fission gas into the oxide is neglected and a uniform temperature is assumed.

The differential equation for the diffusion process is

$$D \frac{d^2c}{dr^2} + \frac{2D}{r} \frac{dc}{dr} + B' - \lambda c = 0 \quad , \quad (1)$$

where

D = Diffusion constant of the fission gas, cm²/sec

c = Number of diffusing atoms/cc

r = Distance from center of sphere, cm

d = Radius of sphere, cm

B' = Rate of formation of fission gas atoms, atom/(cm³)(sec)

λ = Decay constant of fission gas, sec⁻¹

The solution to Equation (1) is

$$c = \frac{B'}{\lambda} - \frac{B'a \sinh(r \sqrt{\lambda/D})}{r \lambda \sinh(a \sqrt{\lambda/D})} \quad (2)$$

Next, we apply Fick's law of diffusion, which states that the rate of transfer of atoms across a unit area is proportional to the concentration gradient, or

$$\frac{\partial m}{\partial t} = -D \frac{dc}{dr} \quad , \quad (3)$$

where m is the number of atoms diffusing across the spherical shell per unit area.

Differentiating Equation (2) with respect to r and evaluating at $r = a$,

$$-\left. \frac{\partial m}{\partial t} \right|_{r=a} = -B' \sqrt{\frac{D}{\lambda}} \left(\coth a \sqrt{\frac{\lambda}{D}} - \frac{1}{a} \sqrt{\frac{D}{\lambda}} \right) \quad (4)$$

Introducing Booth's D' , which is defined as

$$D' = D/a^2 \quad ,$$

Equation (4) becomes

$$-\left. \frac{\partial m}{\partial t} \right|_{r=a} = -B'a \sqrt{\frac{D'}{\lambda}} \left(\coth \sqrt{\frac{\lambda}{D'}} - \sqrt{\frac{D'}{\lambda}} \right) \quad (5)$$

For values of $\lambda/D' > 100$, Equation (5) may be approximated by

$$-\left. \frac{\partial m}{\partial t} \right|_{r=a} = -B'a \sqrt{\frac{D'}{\lambda}} \left(1 - \sqrt{\frac{D'}{\lambda}} \right)$$

or

$$-\left. \frac{\partial m}{\partial t} \right|_{r=a} = -B'a \sqrt{\frac{D'}{\lambda}} \quad (6)$$

This approximation is valid for the fission gases dealt with in this report. The longest half-life considered is the 5.3-day Xe^{133} ($\lambda = 1.52 \times 10^{-6} \text{ sec}^{-1}$). Booth has shown that at 1400°C for UO_2 compacts of 90% theoretical density, D' is of the order of 10^{-9} or $10^{-10} \text{ sec}^{-1}$. In these experiments the value of D' in ThO_2 is not expected to be very different and the temperatures are lower.

Thus the rate of evolution of fission gas from the sphere, in atoms/sec, is

$$\frac{dn}{dt} = +S \frac{\partial m}{\partial t} \quad , \quad (7)$$

where

$$S = \text{Surface area of sphere } (S = 4\pi a^2).$$

Thus

$$\frac{dn}{dt} = 3B' \left(\frac{4\pi}{3} a^3 \right) \sqrt{\frac{D'}{\lambda}} = 3B'v \sqrt{\frac{D'}{\lambda}} \quad ,$$

where ν is the volume of the sphere in cm^3 .

Assuming that the sinter compact is an agglomeration of spheres and summing up over all spheres,

$$\frac{dN}{dt} = 3B' \sqrt{\frac{D'}{\lambda}} \Sigma \nu$$

or

$$\frac{dN}{dt} = 3B \sqrt{\frac{D'}{\lambda}}$$

where B is the rate of formation of gas atoms in the whole compact.

Now dN/dt , the rate at which fission gas atoms escape from the sinter, is expressed as R (atoms/sec) in the text of this report; B , the rate of production of gas atoms in the oxide, may be expressed as FY , where F is the number of fissions occurring per second and Y is the fission yield. Hence,

$$R = 3FY \sqrt{D'/\lambda} \quad (8)$$

or

$$\frac{R}{Y} = 3F \sqrt{D'} : \frac{1}{\sqrt{\lambda}}$$

Hence a plot of R/Y vs $1/\sqrt{\lambda}$ should be linear.

2. Correction for Neutron Capture

In this treatment any removal of the fission gas atoms by neutron capture has been neglected. If this process is significant (as for Xe^{135}), λ in Equation (1) must be replaced by $\lambda + \sigma\phi$, where σ is the capture cross section and ϕ is the thermal neutron flux. Then Equation (8) becomes

$$R = 3FY \sqrt{D'/\lambda'}$$

where

$$\lambda' = \lambda + \sigma\phi$$

3. Correction for Effect of Sheath

Only an unsheathed fuel compact has been considered so far. If a sheath in which there is a defect, is placed around the compact, a second diffusion phenomenon, that of the fission gas in the space between the fuel and the sheath, must be considered.

If the time that a fission gas atom spends in this space is small compared with its half-life, then diffusion from the oxide will be the rate-controlling process and the effect of the sheath will be negligible. However, for a given residence time, as the half-life of a fission gas atom decreases, the sheath will have a more important effect and the release rate of gas from the defect will be less than the expected from Equation (8). There will be some half-life below which no gas will escape, all atoms having decayed before they reach the defect.

The plot of R/Y vs $1/\sqrt{\lambda}$ would then appear as follows:

From the data of Fig. 5-1 of the text, this effect is not apparent and would indicate that the residence "half-life" for a gas atom in the sheath is more probably measured in minutes than in hours.

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