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ARGONNE NATIONAL LABORATORY
P. O. Box 299
Lemont, Illinois

FUEL DEFECT TEST - BORAX-IV

by
R. F. S. Robertson* and V. C. Hall, Jr.

Reactor Engineering Division

*Atomic Energy of Canada, Ltd., Chalk River, Ontario

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TABLE OF CONTENTS

TABLE OF CONTENTS

LIST OF TABLES

LIST OF FIGURES

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ABSTRACT

On March 11 and 12, 1958, the BORAX-IV reactor was operated at a power of 2.4 Mw, even though a large number of the fuel elements contained defects through the cladding. Subsequent investigation showed that 22 of the fuel elements in the reactor contained one or more ruptured rods.

During operation, radiation fields in the vicinity of the turbine and condenser were high, but on shutdown all radiation levels decayed rapidly with a 30-min half-life and after 12 hr all parts of the turbine and condenser showed only normal radiation levels of a few mr/hr.

The effluent gases, the reactor steam, the condensate, and the reactor water were sampled during steady operation and analyzed for fission product gases and other fission products.

From these analyses the following facts were deduced:

(1) The most significant fission products released were fission product gases.

(2) Fission products which had no fission gas as a precursor, such as the radioiodines or Mo^{99}, were not released from the fuel to a significant extent.

(3) The most significant activities found in the turbine and condenser after operation were Ba^{140}-La^{140}, Sr^{90}-Y^{90}, Sr^{89} and Cs^{137}. These are all decay products from short-lived fission product gases.

An analysis of the data was made using information obtained from tests of defect ceramic fuel in EBWR, and an attempt was made to predict contamination levels throughout the system. Predicted levels agreed qualitatively with those found and in general the method led to high results.

On the basis of these experiments it can be predicted that a boiling reactor, fueled with ceramic fuel, can operate for long periods of time with many defects through the cladding provided:

(1) The system is free from leaks.

(2) There is enough hold-up volume in the line to the stack that a delay time of at least one hour is realized.

Under these conditions, although activity levels may be high during operation, no significant amount of long-lived contamination, other than fission gases which have stable daughters, will be released to the reactor surroundings.

1. INTRODUCTION

The BORAX-IV reactor is a direct-cycle boiling water reactor power plant with a net electrical output of 2.5 Mw. The BORAX series of experiments has included the use of different core loadings for the study of stability and operating characteristics of boiling water reactors.[1,3]

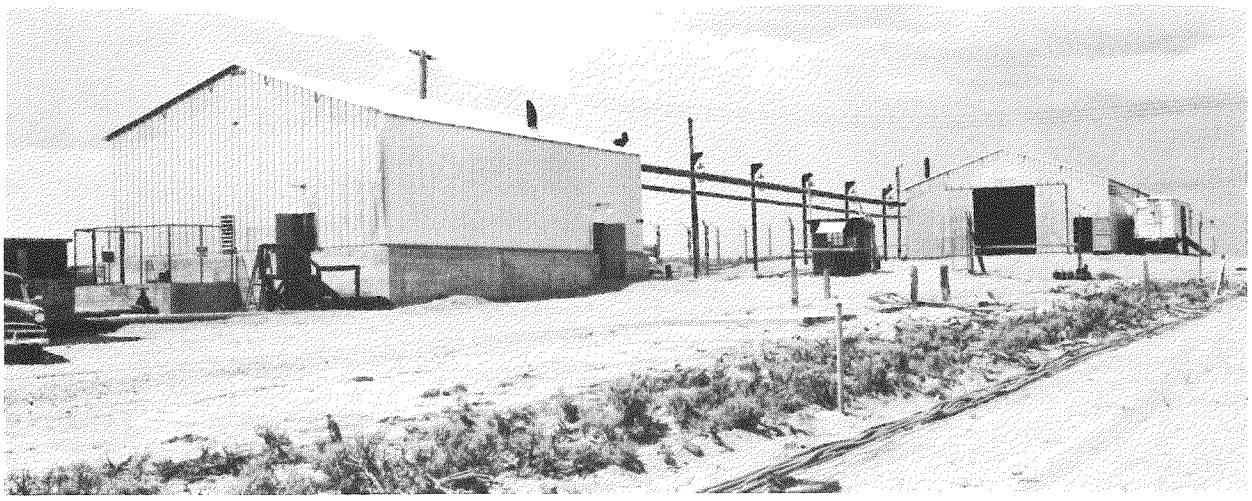
Figure 1-1 is a pictorial plant layout of the BORAX facility. The Control Trailer is located approximately one-half mile away from the reactor plant to permit operation of the plant under adverse conditions.

Figure 1-2 is a simplified flow diagram for the complete plant.

The BORAX-IV fuel elements[2] consist of thoria-urania pellets, bonded with lead in aluminum-1% nickel tube plates. The pellets are $\frac{1}{4}$ in. in diameter and $\frac{1}{2}$ in. long. The pressing mixture for the pellets consisted of 93.4 wt % ThO_2 and 6.54 wt % U_3O_8. The U_3O_8 was above 90% enrichment. The pellets were fired in air at 1700-1750° C, attaining an average geometric density of 8.3 g/cc and an average apparent density of 9.8 g/cc. The effective cladding thickness is 0.016 in., while the effective thickness of the lead sealer is 0.018 in.

The last power run with the 59-element BORAX-IV core was made on December 5, 1957. Following this run the core was revised so as to increase the maximum power for continuation of stability studies.[3] The core revision included the removal of the boron-steel flux-depression wires from the 16 central elements as well as the addition of 10 new elements to the periphery of the core.

The reactor was started at 1000 on the morning of February 19, 1958. At 2100 the power was increased from 500 kw to 2.4 Mw and the turbine was started. At 5 Mw (thermal) the hotwell activity was 700 mr/hr. At 10 Mw (thermal) the activity increased to 9 r/hr (approximately five times the maximum previously observed at this power level). At this time considerable steam was coming from the reactor pump pit and exhaust stack, and the reactor was shut down. The following day two of the electrical startup heaters were found to be ruptured. It was assumed that the high level of activity was caused by the MgO and other impurities which were released into the reactor water when the Calrods ruptured. The heaters were subsequently repaired.



TURBINE (LEFT) AND REACTOR (RIGHT) BUILDINGS
CABLES IN FOREGROUND LEAD TO CONTROL TRAILER ONE-HALF MILE DISTANT

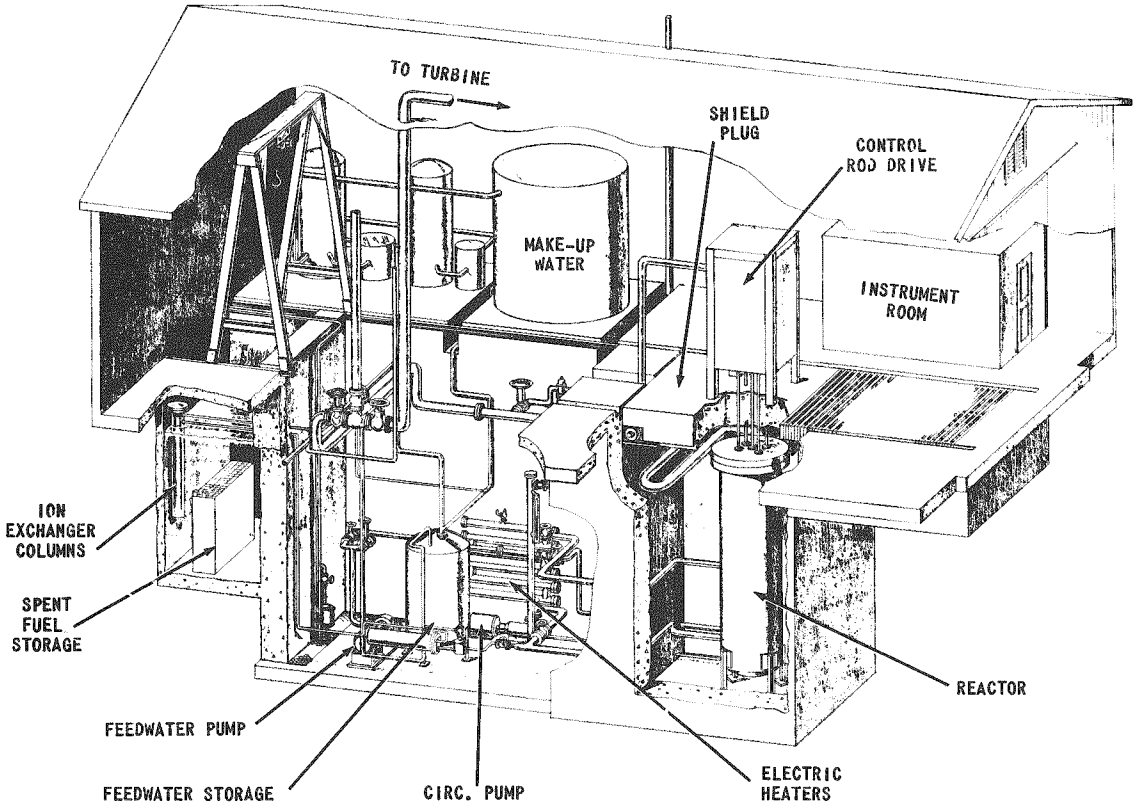
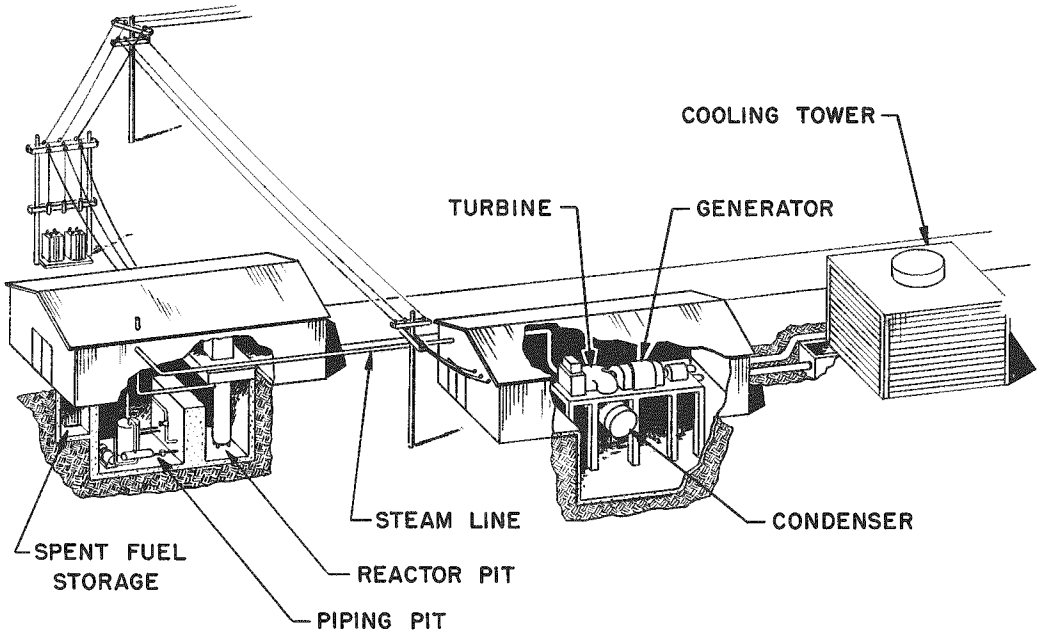


FIG. 1-1
REACTOR AND TURBINE BUILDINGS



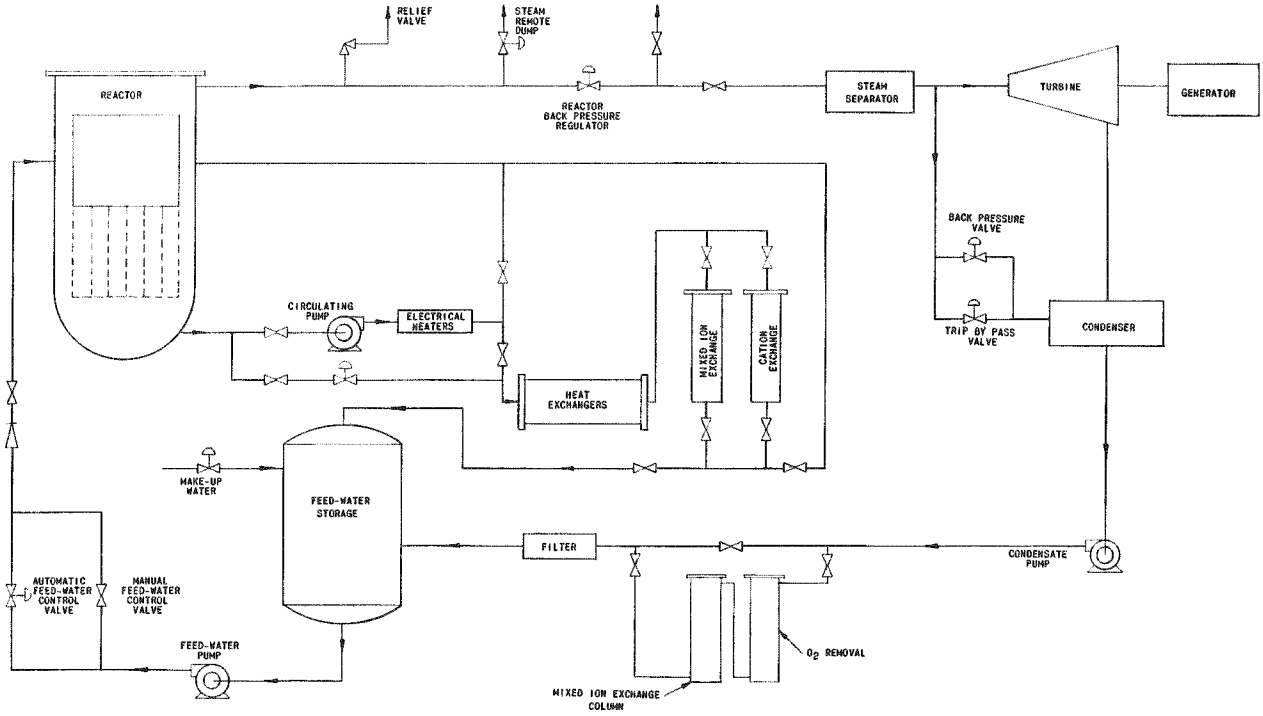


FIG. 1-2
SIMPLIFIED FLOW DIAGRAM

On February 25, another power run was attempted. At 12 Mw (thermal) the condenser hotwell activity was 8 r/hr. The reactor was shut down and the activity decayed with a 33-min half-life to normal background levels. The principal source of activity was identified as Cs^{138}, by both its half-life and gamma-energy spectrum.

It was concluded at this time that a fuel rupture had occurred. The extent of the rupture and quantitative effects were unknown. The rapid decay to background activity levels indicated that the reactor could be operated for further evaluation.

On March 11 and 12, 1958, the reactor was operated at low power as a special experiment to evaluate the effect of running a direct-cycle boiling water reactor with a fuel element defect. The remainder of this report deals with the results obtained during these two days of operation.

2. OBJECTIVES

The principal objective of this experiment was to determine the limiting factors on the operation of this reactor with a fuel defect. Measurements which were made during the reactor operation included the following:

a. radioactivity levels of the steam plant equipment;
b. quantitative determination of the fission gases Xe^{138} and Kr^{88}, which were released through the air ejector;
c. analyses of reactor water, condensed steam before the turbine, and condensed steam after the turbine (hotwell condensate) for fission products; and
d. area contamination downwind from the reactor.

3. EXPERIMENTAL METHODS

3.1 Reactor Operation

The plot of reactor operation for the two-day test period is shown in Fig. 3-1.

At room temperature the reactor loading required that approximately 1350 gm of boric acid be present in the water to maintain the reactor in a subcritical condition. Thus, about three hours were required initially to preheat the vessel and remove the boric acid on the ion-exchange resins. As soon as the poison was removed to a suitable extent (specific conductivity of water = 5 micromhos-cm^{-1}) the flow to the ion-exchange was shut off.

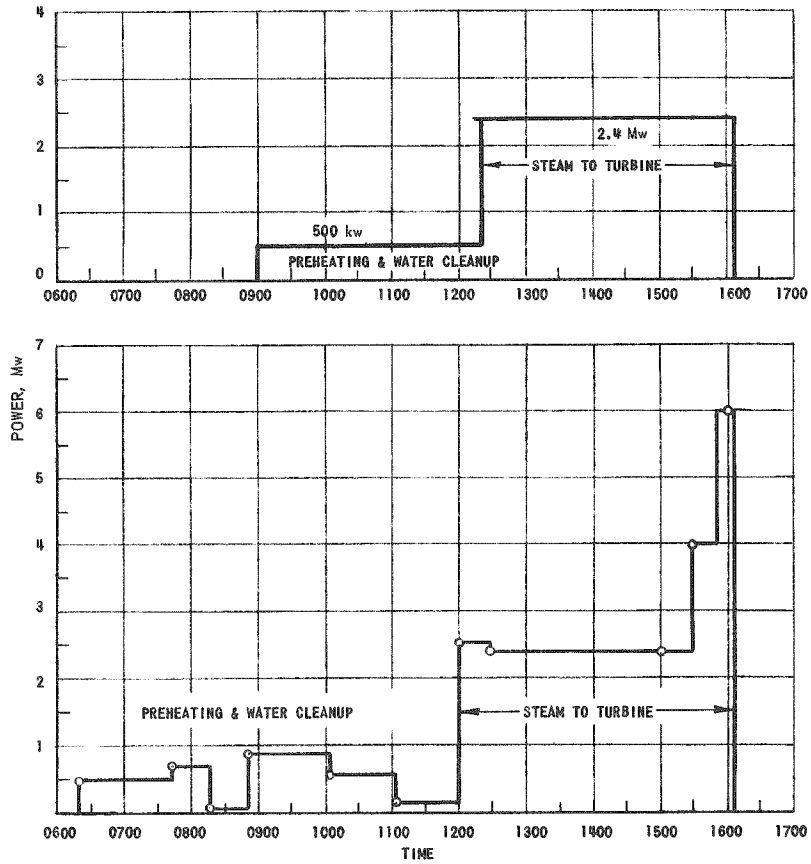


FIG. 3-1
BORAX IV OPERATION

The power level of 2.4 Mw(t) was chosen as the lowest which could be maintained and yet run the entire plant under equilibrium conditions. During both the tests the reactor was run only long enough to achieve thermal equilibrium conditions and to take the necessary samples, after which it was shut down.

3.2 Special Precautions

During these tests the radioactivity levels around the reactor building and the turbine building were high because of direct radiation and because of release of radioactive fission products through leaks.

All persons taking samples or conducting radiation surveys were required to change to protective clothing. Scott Air Paks were used throughout. Before leaving the reactor area all personnel were checked at a monitoring station.

3.3 Radiation Surveys

Radiation surveys around the reactor and turbine buildings were carried out with portable Juno Survey Meters.

In addition, high-volume air-sampling stations were set up downwind from the reactor area at distances of 150 and 500 ft.

3.4 Samples and Analyses

During the two runs, the reactor water, the steam, the condensate in the condenser hotwell, and the air ejector gases were sampled and analyzed for fission product radionuclides.

Analyses for radionuclides in the reactor water, condensed steam, and hotwell condensate were carried out by conventional radiochemical methods.

The samples of gases emitted from the air ejector were analyzed[4] for Xe^{138} and Kr^{88}. Representative samples of the gas were taken in bulbs of known volume at a known pressure. The bulb was allowed to stand for a known time (roughly $1\frac{1}{2}$ hr) and the contained gases were then allowed to expand into another evacuated bulb of known volume. The Cs^{138}, which had collected on the walls of the first bulb after formation through Xe^{138} decay with a half-life of 17 min, was washed off and counted in a proportional counter. The identity of the Cs^{138} was established by following the decay of the activity (33-min-half-life) and by gamma spectrometry. The second bulb was allowed to stand a further $1\frac{1}{2}$ hr and then the Rb^{88}, which had accumulated after formation from the decay of Kr^{88} with a half-life of 2.8 hr, was washed out and counted. Its identity was determined by following the decay curve (18-min half-life). Knowing the times involved, the activities of Xe^{138} and Kr^{88} in the original sample could be determined. From this and a knowledge of the flow rate of the gases through the air ejector, the rate of emission of Xe^{138} and Kr^{88} could be calculated.

3.5 Metal Coupons in Turbine

Before reactor operation on March 11, the manhole cover of the turbine exhaust casing was opened and coupons of various metals were inserted in a special holder. These coupons were discs, one inch in diameter, which were made from mild steel, stainless steel, aluminum, and copper. The exhaust casing was closed and the samples were left in place during reactor operation on March 11 and 12. On March 13 they were removed and were taken to ANL, where they were examined for gamma-ray spectrum in a 256-channel gamma-ray pulse-height analyzer.

4. EXPERIMENTAL RESULTS

4.1 Radiation Levels

4.1.1 Equipment Surfaces

Table 4-1 lists the radiation levels at various locations in the system at steady operation. The last column shows the relatively low activity obtained during normal operation at 12 Mw when it was known that no defects were present. These levels may be attributed primarily to N^{16} [$O^{16}(n,p)N^{16}$], since on reactor shutdown they decayed with a 7-sec half-life.

Table 4-1

RADIATION LEVELS DURING OPERATION (mr/hr)

Location	With Defect 2.4 Mw	With Defect 6.0 Mw	No Defect 12 Mw
Reactor Building			
Main Steam Line	500	500	250
Feed-water Filter	6,000	12,500	< 50
Top of Reactor	160	210	<100
Main Door to Building	20	30	< 10
Turbine Building			
Main Steam Line	250	400	200
Turbine Exhaust Casing	150	190	50
Air Ejector after Cooler	15,000	-	420
Air Ejector Exhaust Filter (A-C Type)	30,000	50,000	Not in Service
Hotwell	2,200	-	500
Main Door to Area	20	27	< 10

It is obvious that during operation on March 11 and 12, radiation levels everywhere were significantly higher than normal. On reactor shutdown they no longer fell off rapidly, but decayed with a half-life of roughly 30 min. However, after a period of 17 hr all radiation levels had decayed to their initial low values.

Measurements of internal radiation levels in the turbine exhaust casing showed that no activity had built up during this experiment.

4.1.2 Downwind Activities

Six 25-cfm air samplers were in operation downwind from the Air Ejector Exhaust Stack. Three samplers were placed on an arc 125 ft downwind with a spacing of 20 ft; three samplers were placed on an arc 500 ft downwind with a spacing of 125 ft. The air samplers work on the

principle of passing air through a filter paper at a known rate and measuring the resultant activity on the filter paper. In addition, a field survey was made along the downwind centerline with a Geiger-Mueller counter. Measurements of the filter paper from the air samplers and the field survey were made approximately one hour after reactor shutdown. The observed activities decayed with a half-life of 33 min. The values shown in Table 4-2 have been calculated back to time of reactor shutdown on the basis of the 33-min half-life.

Table 4-2

FIELD MONITORING - MARCH 12, 1959

High-volume Air Samplers

Location	Activity
125 ft Downwind	350 mr/hr (average of 3 samplers)
500 ft Downwind	150 mr/hr (average of 3 samplers)

Field Survey

Along Downwind Centerline

(Normal Background, 0.03 mr/hr)

Distance from Gas Discharge Stack (ft)	Ground Surface Activities, β and γ (mr/hr on contact)
125	4.0
200	2.7
300	1.6
500	3.0
750	1.8
1000	0.12

4.2 Rates of Xe^{138} and Kr^{88} Release from Defective Fuel Elements

Table 4-3 shows results obtained. It can be seen that the agreement between samples was fair.

4.3 Radioactivities in Reactor Steam, Hotwell, and Reactor Water

Samples of reactor steam, reactor water, and hotwell condensate were taken during each period of operation at 2.4 Mw. Results of analyses are shown in Table 4-4.

Table 4-3

RATES OF RELEASE OF Xe^{138} AND Kr^{88} FROM DEFECTIVE FUEL ELEMENTS

Sample No.	Date	Time	Bulb Volume (ml)	Activity of Gas, 10^8 d/sec/l(STP) of Gas		Gas Flow Rate, l(STP)/min	Emission Rate (Curies/min)	
				Xe^{138}	Kr^{88}		Xe^{138}	Kr^{88}
7-2	3-11-58	1415	300	2.32 5.7	1.23[a]	183		
7-4	3-11-58	1415	17	3.8 8.5	1.82[a]	183		
17-1	3-12-58	1400	21	7.3	1.2[a]	183		
17-2	3-12-58	1415	14.8	12.3	1.2[a]	183		
		Average:		(8 ± 4)	(1.4 ± 0.2)		4.0 ± 2[b]	0.7 ± 0.1

[a]Average of two analyses which agreed within 10%.

[b]Omitting low values for Sample No. 7-2.

Table 4-4

RADIOACTIVITIES IN REACTOR WATER, STEAM, AND CONDENSATE

All activities expressed as disintegrations per min per ml of water or condensed steam
All activities corrected to sampling time

Date	Time	Location	Cs^{138}	Cs^{137}	Ba^{140}	Sr^{89}	Sr^{90}	I^{131}	Mo^{99}
3-11-58	1107	Reactor Water						710	
3-11-58	1410	Reactor Water	2.2×10^6	1.1×10^4	4.3×10^3	1.1×10^4	420	4.0×10^3	
3-11-58	1508	Steam		19	120	1.2×10^3	neg	120	
3-11-58	1415	Condensate	2.2×10^6	16	60	570	neg	130	
3-12-58	1330	Reactor Water	1.3×10^6	1.2×10^4	7.5×10^3	1.7×10^4	484	3.1×10^3	1.6×10^4
3-12-58	1323	Steam	1.2×10^7	20	120	690	neg	90	neg
3-12-58	1325	Condensate	1.6×10^6	24	47	590	neg	70	neg

N.B. (1) "Neg" means <10 d/min/ml.

(2) Uncertainty in above results in about ±30%.

Aliquots of samples from the same sources were also counted on a gas flow proportional counter and the decay was followed with time. Decay curves are shown in Fig. 4-1. Figure 4-2 shows the initial, short-lived decay of the steam and condensate samples.

Examination of the decay curves shows an interesting correlation with the radiochemical data.

The Cs^{138} activity is considered first. Figure 4-2 shows that in all steam samples the activity decayed initially with a 33-min half-life, indicating that the preponderant activity was Cs^{138}. The efficiency of the

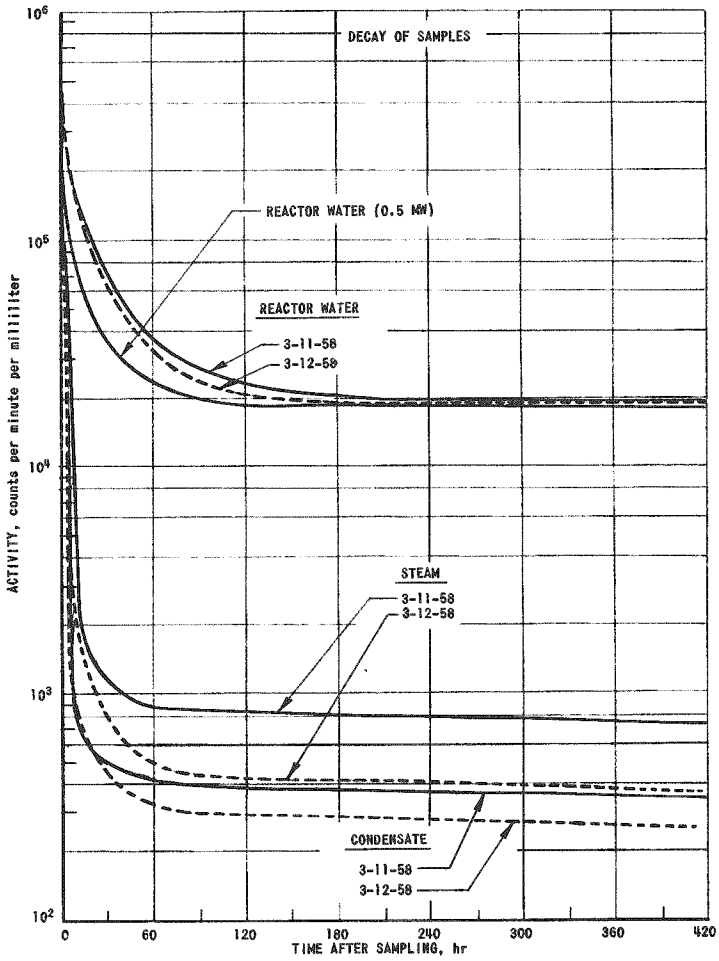


FIG. 4-1
DECAY OF ACTIVITY IN WATER, STEAM, AND CONDENSATE SAMPLES

counter used was found experimentally to be 33%. Extrapolating the 33-min activity back to sampling time, and using this efficiency, the Cs^{138} activity was calculated. The Cs^{138} activity from Fig. 4-2 is compared in Table 4-5 with that found by chemical separation and counting (see Table 4-4).

The longer-lived activities are next considered. Gamma-ray spectra were measured on samples of reactor water about 24 days after sampling. Gamma rays of the following energies were present:

Energy (Mev)	Nuclide	Energy (Mev)	Nuclide
2.5	La^{140}	0.78	Co^{58}
2.1	La^{140}	0.66	Cs^{137}
1.6	La^{140}	0.49	La^{140}, Co^{58}
0.9	Y^{89}	0.33	La^{140}, I^{131}

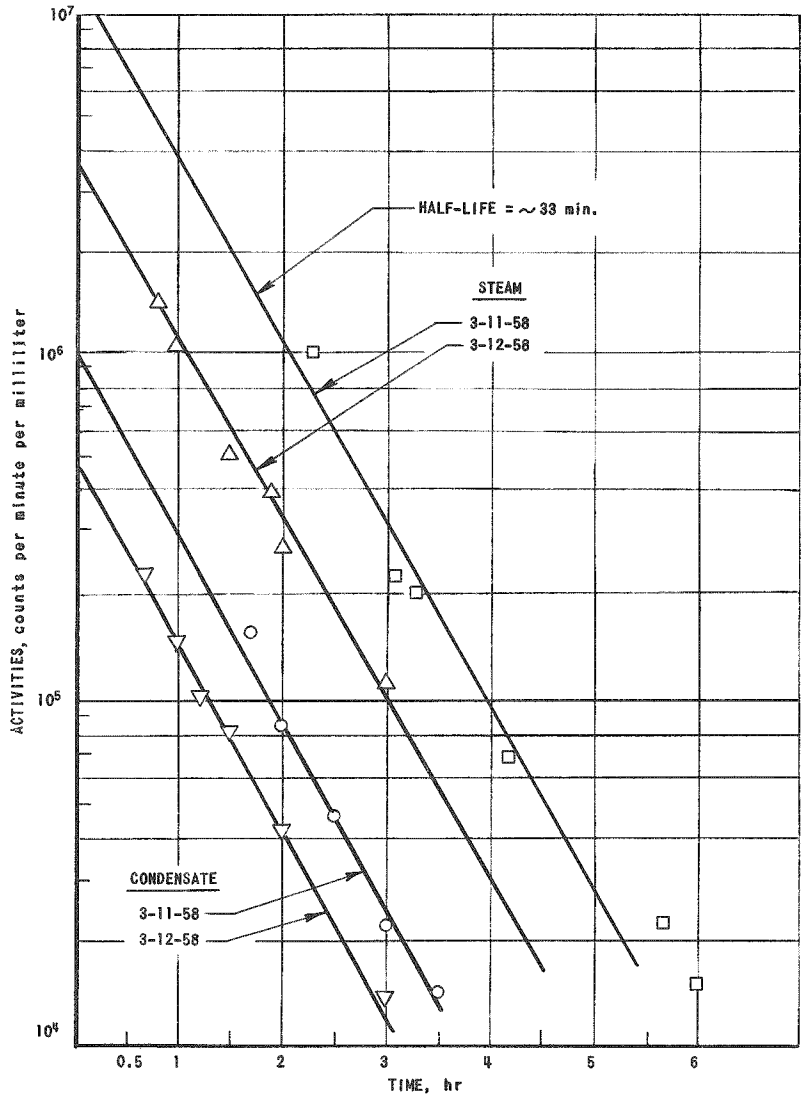


FIG. 4-2
INITIAL SHORT-LIVED DECAY OF STEAM SAMPLES

Table 4-5

COMPARISON OF Cs^{138} ACTIVITIES FROM TABLE 4-4 AND FIG. 4-2 ACTIVITIES IN d/(min)(ml)

Time	Date	Location	From Table 4-4	Calculated from Fig. 4-2
1508	3-11-58	Steam		3.9×10^7
1323	3-12-58	Steam	1.2×10^7	1.2×10^7
1415	3-11-58	Condensate	2.2×10^6	3.3×10^6
1325	3-12-58	Condensate	1.6×10^6	1.5×10^6

The La^{140} will have been in radiochemical equilibrium with Ba^{140} at the time of counting. It is estimated that at this time these γ rays accounted for well over 90% of the observed γ activity.

Ba^{140} activities could be calculated from the observed La^{140} activities, and at reactor shutdown the following ratio of disintegration rates could be calculated:

$$Cs^{137} : Ba^{140} : Co^{58} = 2:1:0.6 \quad .$$

The Cs^{137} : Ba^{140} ratio agrees reasonably well with that found from Table 4-4.

The Co^{58} is presumed to come from the nickel present in the aluminum-nickel sheathing by the fast neutron reaction $Ni^{58}(n,p)Co^{58}$.

Sr^{89} and Sr^{90} are also present in the reactor water, the former to a significant extent, but since neither of these nuclides emit γ rays they are not detected by γ counting.

Thus, the nuclides listed in Table 4-4 and Co^{58} contribute well over 90% of the observed gamma activity in the reactor water and the majority of the beta activity.

Gamma-ray spectra were also measured on condensed steam samples taken from the steam line 24 days after sampling. The following gamma-ray energies were present:

Energy (Mev)	Nuclide
1.6	La^{140}
0.92	Y^{89m}
0.82	La^{140}
0.66	Cs^{137}
0.49	La^{140}
0.33	$La^{140}(I^{131}?)$
0.17	Ba^{140}

Thus, with the exception of Co^{58} the same nuclides were present, but their ratios were quite different. In these samples the ratio of the disintegration rates was

$$Cs^{137} : Ba^{140} = 1:14 \quad .$$

This ratio is somewhat larger than that indicated in Table 4-4, but the values are in qualitative agreement. The very low counting rate in these samples made quantitative estimation of activity from the γ-ray peaks difficult.

The nuclides listed above again accounted for over 90% of the observed γ-ray activity. That the nuclides listed in Table 4-4 account for the majority of the beta activity can be shown as follows:

The reactor steam sample taken on March 12 will be used as an example. The decay curve for this sample shows that, about 300 hr after sampling, the beta-counting rate was about 400 c/(min)(ml). The efficiency of the counter used was about 33%; hence the beta disintegration rate was about 1200 d/(min)(ml). Now, the disintegration rates shown in Table 4-4 for nuclides in this sample can be extrapolated to 300 hr after sampling and the individual rates summed. The fact that La^{140}-Ba^{140} and Y^{90}-Sr^{90} will be in radiochemical equilibrium must be taken into account. The total disintegration rate is then roughly 900d/(min)(ml). In other words the nuclides of Table 4-4 account for 75% of all beta disintegrations. This figure is probably conservative; for instance, if the efficiency of the counter were actually 40%, then 90% of the beta disintegrations would have been accounted for.

4.4 Radioactivities on Metal Coupons

The γ-ray spectra of the various metal coupons removed from the turbine exhaust casing were very similar to the spectra of the reactor steam samples and showed the presence of only Ba^{140}, La^{140}, and Cs^{137}. These three nuclides contributed over 95% of the observed γ-ray activity deposited on the coupons.

It must be remembered that Sr^{89} and Sr^{90} have no γ rays and hence would not be detected by γ-ray pulse-height analyses. No radiochemical separations or beta counting was done on the coupons; hence the presence of these two nuclides on the coupons can only be presumed.

From the 1.6-Mev γ-ray peak of La^{140} and the 0.66-Mev γ-ray peak of Cs^{137}, the disintegration rates of these two nuclides could be computed. The Ba^{140} peaks, although suitable for definite qualitative identification, were not suitable for quantitative interpretation. Ba^{140} activities could be calculated, however, on the assumption that at the time of reactor shutdown on March 12, La^{140} activities were low and that the La^{140} activity observed at counting time resulted from decay of Ba^{140}.

Results are shown in Table 4-6. Activities of the three nuclides are seen to be very low.

The absolute counting rates shown in Table 4-6 are uncertain to about ±50%, but the ratios of a given nuclide from one metal specimen to another are much more accurate; the uncertainty is less than ±10%.

Table 4-6

RADIOACTIVITIES ON METAL COUPONS

Metal	Activity/Coupon 137 hr after Reactor Shutdown (disintegrations/min)			Activity at Reactor Shutdown March 12, 1958 (μcurie/sq ft)		
	Ba^{140}	La^{140}	Cs^{137}	Ba^{140}	La^{140}	Cs^{137}
Mild Steel		7.4×10^3	775	0.42		0.032
Stainless Steel		6.6×10^3	890	0.37		0.037
Aluminum		5×10^3	635	0.28		0.026
Copper		4×10^3	510	0.22		0.021

The amount of activity deposited on a given surface will undoubtedly depend on a number of factors; the physical and the chemical nature of the surface will play an important role as will the velocity of the carrier steam past the sample.

From this experiment the importance of these factors cannot be assessed. However, the general conclusion may be drawn that, if the activity deposition on these specimens was representative of deposition in the turbine and condenser as a whole, the reactor operation on March 11 and 12 led to very low contamination by these nuclides.

This subject will be discussed further in Section 7.3.2.

5. CALCULATIONS

5.1 General

In these calculations and throughout the remainder of this report, the following parameters and constants will be used:

Reactor Parameters

Power (thermal)	2.4 Mw
Wt U^{235} in core	19.5 kg
Steam Flow at 2.4 Mw	7.6×10^3 lb/hr = 958 gm/sec
Volume of Water in Reactor	1000 gal (at 25°C) = 3.79×10^3 liters

If it is assumed that for each fission 190 Mev appear as thermal energy in the water, then

$$1 \text{ Mw} = 3.3 \times 10^{16} \text{ fissions/sec}$$

and from this, the average fission rate F in the reactor at 2.4 Mw is

$$F = (3.3 \times 10^{16} / 19.5 \times 10^3)\ 2.4$$

$$= 4.06 \times 10^{12} \text{ fissions/(sec)(gm } U^{235}) \quad .$$

The data listed in Table 5-1 will also be used.

Table 5-1

NUCLEAR DATA

Nuclide	Half-life	Decay Constant λ (sec^{-1})	Fission Yield Y (%)
Kr^{88}	2.78 h	6.93×10^{-5}	3.6
Rb^{88}	18 m	6.42×10^{-4}	3.6
Kr^{89}	2.6 m	4.44×10^{-3}	4.8
Sr^{89}	54 d	1.57×10^{-7}	4.8
Kr^{90}	33 s	2.1×10^{-2}	5.8
Sr^{90}	28 y	7.85×10^{-10}	5.8
Xe^{137}	3.8 m	3.04×10^{-3}	6.1
Cs^{137}	30 y	7.33×10^{-10}	6.1
Xe^{138}	17 m	6.80×10^{-4}	5.7
Cs^{138}	33 m	3.50×10^{-4}	5.7
Xe^{140}	16 s	4.33×10^{-2}	6.4
Ba^{140}	12.8 d	6.27×10^{-7}	6.4
I^{131}	8.07 d	9.94×10^{-7}	2.9
Mo^{99}	66 h	2.92×10^{-6}	6.1

5.2 Calculation of U^{235} Equivalence of Fission Gas Release Rates

The mass of U^{235} which must be exposed to the water to release fission product gases at the observed rate was calculated, based upon the following assumptions:

a. Fission fragments enter the water as fission recoils from a U^{235}, or more probably a $U^{235}O_2$, surface, which is exposed to the water. The range of a fission fragment in ThO_2 is of the order of 5 to 8 x 10^{-4} cm. Thus a fragment born in the oxide lattice farther than this from the water surface will not reach the water, and also only a certain fraction (25%) born within this thin strip will reach the water. Thus, any calculation will represent only the minimum amount of U^{235} which could produce the observed quantity of gas.

b. A recoil fission fragment may be a precursor of the fission gas in a particular fission chain. It is assumed that these precursors (Se, Br, Te, or I) remain in the reactor water, or on metal surfaces in the water. This is undoubtedly true in the case of Se or Te, but a small amount of radiobromines or radioiodines may be stripped from the water. Table 4-4 shows that the I^{131} activity in the steam was only about 3% of that in the water. Thus the assumption of no stripping of radioiodines or bromines is considered good.

c. Any fission product gases produced are stripped completely to the steam phase, and the delay time from the steam dome of the reactor to the sampling point in the steam line or to the air ejectors is short compared to the half-life of the gas.

This assumption of complete stripping may be checked by considering the measured activities of Cs^{138}. Table 4-3 shows that Xe^{138} was released at a rate of 4 curies/min. This corresponds to a rate of

$$\frac{4 \times 3.7 \times 10^{10}}{60 \times 6.8 \times 10^{-4}} = 3.63 \times 10^{12} \text{ atoms/sec} \quad .$$

Thus, using the assumption outlined, the release rate of Xe^{138} to the reactor water must be 3.63 x 10^{12} atoms/sec.

Now suppose it is assumed that Xe^{138} is released to the reactor water at this rate but none is stripped out, i.e., it all remains in solution, decaying there to Cs^{138}. Then the Cs^{138} equilibrium activity may be calculated to be 5.7 x 10^7 d/(min)(ml). However, the measured activity was only 2 x 10^6 d/(min)(ml) (Table 4-4), i.e., only about 3% of the calculated value. Thus Xe^{138} must have been removed extensively from solution.

The postulate of complete stripping can be checked by another calculation. During its passage via the steam pipe to the turbine and condenser, Xe^{138} will be decaying to Cs^{138}. Some of this Cs^{138} will remain on equipment surfaces but some will be washed down to the condenser hotwell and returned thence via the feedwater to the reactor vessel. Now it will be assumed that Xe^{138} is completely stripped from the reactor water, and that the only means by which Cs^{138} can appear is via the feedwater.

Table 4-4 shows that the condensate activity in the hotwell was about 2×10^6 d/(min)(ml). At a flow rate of 958 gm/sec, Cs^{138} will be returning to the reactor at a rate of 9.14×10^{10} atoms/sec. The delay time between the hotwell and the reactor is about 5 min, which is small compared to the 33-min half-life of Cs^{138}. Thus the Cs^{138} will be appearing in the reactor water at a rate of 9.14×10^{10} atoms/sec and will be decaying at a rate of λN disintegrations/sec, where N is the total number of Cs^{138} atoms in the reactor water. At steady state, then, the Cs^{138} activity will be given by

$$\frac{9.14 \times 10^{10} \times 60}{3.79 \times 10^6} = 1.4 \times 10^6 \text{ d/(min)(ml)} \quad .$$

This is in reasonable agreement with the measured value of 2×10^6 d/(min)(ml) and indicates that the assumption of complete stripping of the fission gas is justified.

If the foregoing assumptions are made together with the assumption that the reactor has operated for a sufficiently long period of time for the fission gas precursors to be in radiochemical equilibrium, then the rate of production, and hence rate of emission of a fission product gas, will be given by:

$$\text{Production rate } R = M^{235} FY \text{ (atoms/sec)} \quad , \qquad (5.1)$$

where

M^{235} is the mass of U^{235} in grams

F is the fission rate per gram of U^{235}[fission/(sec)(g U^{235})]

Y is the fission yield

R is an experimentally measured value.

This is, of course, an oversimplified expression which ignores burnup of the U^{235} and assumes that fission products come only from U^{235} fission. It is accurate enough for the purposes of this report. The quantities F and Y are known; hence M^{235} may be calculated. Table 5-2 shows the results for Xe^{138} and Kr^{88}.

Table 5-2

CALCULATION OF MINIMUM U^{235} EXPOSED

Nuclide	Curie/min	Atom/sec	(R/Y) x 10^{13}	M^{235} (gm)
Xe^{138}	4.0 ± 2.0	(3.6 ± 1.8) x 10^{12}	(6.3 ± 3.2)	15.5 ± 7
Kr^{88}	0.7 ± 0.1	(6.2 ± 0.9) x 10^{12}	(17 ± 3)	42.5 ± 6.5

5.3 U^{235} Equivalence of Other Fission Products

The U^{235} equivalences of radionuclides in the reactor water are calculated by a method similar to that used above.

For a given nuclide,

$$\frac{dN}{dt} = M^{235}\,FY - \lambda N \quad , \tag{5.2}$$

where

N is the total number of nuclide atoms in the reactor water.

Any cleanup by the ion-exchange resin can be ignored, since the flow to the resin columns was valved off before the reactor came to power. Integration of Eq.(5.2) with the boundary condition N = 0 when t = 0, gives

$$\lambda N = M^{235}\,FY\,(1 - e^{-\lambda t}) \quad . \tag{5.3}$$

In all cases the time interval t, the time between reaching 2.4 Mw power and the time the sample is taken, was about 1.5 hr and was thus short compared with the half-life of any nuclide in question. Thus, to a good approximation,

$$\lambda N = M^{235}\,FY\lambda t \quad . \tag{5.4}$$

Knowing λN, the total activity of a given nuclide in the reactor water, and knowing F, Y and λ, a value of M^{235} may be calculated.

A value for R, the release rate of the nuclide to the water (atoms/sec), may also be calculated from Eq. (5.1).

Individual nuclides are dealt with below.

5.3.1 I^{131} and Mo^{99}

Table 4-4 shows that at 1107 hr on March 11, when the reactor power was about 0.5 Mw, the I^{131} activity was about 700 d/(min)(ml). At 1410 hr, the I^{131} activity had risen to 4×10^3 d/(min)(ml). It is assumed that in two hours the activity in solution had increased by $(4 - 0.7) \times 10^3$ d/(min)(ml). On this basis, Table 5-3 shows that the U^{235} equivalent to this activity was about 0.25 gm.

Table 5-3

U^{235} EQUIVALENCE OF I^{131} AND Mo^{99} IN REACTOR WATER

Date	Time	Nuclide	Time at Steady Power (2.4 Mw) hr	Initial Activity, d/(min)(ml)	Activity at Sampling Time, d/(min)(ml)	M^{235}, gm	R/Y
3-11-58	1410	I^{131}	2	7×10^2	4×10^3	0.25	1×10^{12}
3-12-58	1330	Mo^{99}	$1\frac{1}{2}$	?	1.6×10^4	0.26	1.1×10^{12}

Table 4-4 shows the Mo^{99} level to be 1.6×10^4 d/(min)(ml) at 1330 on March 12. The initial Mo^{99} activity is not known. It must be remembered that only an anion bed was used to remove H_3BO_3 from the reactor water before each run; hence I^{131} would be removed but probably cationic species would not.

A "worst case" assumption will be made, i.e., that between reactor startup on March 13 and the time of sampling (after about $1\frac{1}{2}$ hr) the Mo^{99} activity rose from zero to 1.6×10^4 d/(min)(ml). The calculated U^{235} equivalent is again about 0.26 gm.

These values of M^{235} are significantly lower than those calculated for the fission gases. The values of R/Y are also very much lower.

5.3.2 Cs^{137}, Sr^{89}, Sr^{90} and Ba^{140}

It has been shown previously that these three nuclides contributed over 50% of the long-lived activity present in the reactor water. They undoubtedly arise from the decay of the shorter-lived Xe and Kr precursors. Some of these precursors, the fraction depending on their half-life, will be removed from the system via the air ejector exhaust system. The remainder will decay to the Cs or Sr which will deposit on equipment surfaces or will be returned to the reactor water via the condenser hotwell and feed-water pumps. It is thus difficult to make any significant calculations regarding these isotopes.

In Section 7.3.2 it will be shown that the activities found agree reasonably well with the postulated mechanism of fission gas release.

The reason for the high Cs^{137}, Sr^{89}, and Ba^{140} activities in the reactor water at the start of the experiment are not known, but presumably they were present before March 11. Unfortunately, no analyses for these nuclides were performed on the sample taken at 1107 on March 11, when the reactor was at a power of 0.5 Mw. The decay curve (Fig. 4-1) indicates that the same long-lived activities were present at this time.

The most probable explanation for the presence of the large amount of these nuclides is that they were emitted as a burst of activity when the defect first occurred, or perhaps as bursts when the reactor was cycled at higher power during the period February 19-25 (see Section 1).

5.4 Correlation of Foregoing Results

The calculations of sections 5.2 and 5.3 have shown that a simple mechanism of release of fission products from fission recoils cannot explain the observed results.

In particular it was found that the only significant radioactivities found were due either to fission gases or their decay products and that no significant amounts of nuclides without gaseous precursor were found.

It was also found that for the function R/Y was not constant but decreased with decreasing half-life of the fission gas.

This behavior is in accordance with that found during tests of defected ceramic fuel in EBWR.[4] In these tests pellets of ThO_2-UO_2 ceramic (enriched in U^{235}) were exposed in EBWR in stainless steel sheaths through which a small hole had been drilled.

When no defect fuel was present in EBWR the values of R/Y were found to be constant for five fission gases: Xe^{138}, Kr^{88}, Kr^{85m}, Xe^{135}, and Xe^{133}. Thus in this case the presence of these gases can be attributed to the presence of fission recoils from a thin film of uranium oxide contamination on the exterior surfaces of the fuel element. The amount of contamination corresponded to the presence of 1 mg of U^{235}.

When a defect sample was present, however, the function R/Y was no longer constant for these five gases. It was found that if R/Y was plotted against $\lambda^{-1/2}$, where λ is the decay constant, a straight line passing through the origin resulted. There was an indication that the radioxenons fell on one line while the radiokryptons fell on another straight line of higher slope.

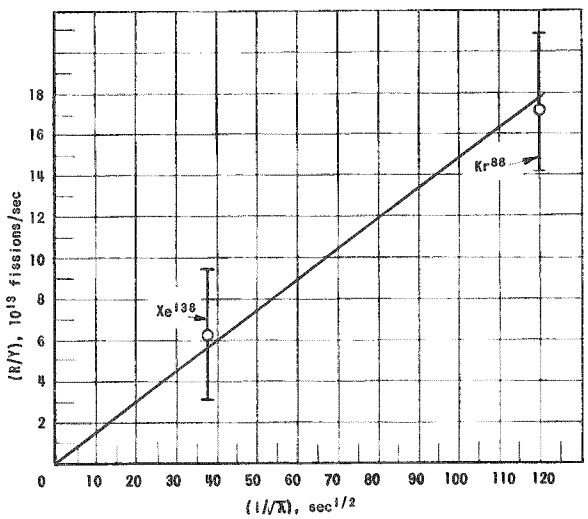


FIG. 5-1
(R/Y) VS $\lambda^{-1/2}$

Figure 5-1 shows that in this experiment the Xe^{138} and Kr^{88} data may be correlated by such a plot, although the uncertainties in this experiment were much greater than those in the EBWR experiments.

This behavior may be explained by some diffusion-controlled mechanism within the fuel element. Whether this diffusion is through the oxide matrix or through the gap between the fuel and the sheath has not yet been established definitely. It is definite, however, that the time delay between the time of production of a fission gas atom within the fuel and time when it reaches the defect is long enough that a large fraction of the very short-lived gaseous nuclides will have decayed before they reach the defect.

Thus, a small defect (or many small defects) must be presumed, behind which there is a reservoir in which gases can diffuse. Subsequent examination of a fuel element (see Section 6) has shown that defects had occurred in the void at the top of the fuel tubes. Thus the desired mechanism is present, all the more so if it is assumed that the lead bond between the pellets and the aluminum-nickel sheath was porous.

The magnitude of the release rate of these gases, however, demands that many such defects were present in the fuel elements. Again, subsequent examination[5] (Section 6) showed that of 69 fuel elements, 22 contained defective plates.

5.5 Decontamination Factors

In a boiling water reactor, concern has always been expressed regarding the extent to which activity will be transferred by the steam to the turbine and condenser. A decontamination factor (DF) may be expressed

as the ratio of the activity per unit weight of reactor water to the activity in the same weight of steam. The DF may be expressed for gross activity or, better, for individual nuclides.

Since fission product gases are so completely stripped from the reactor water, i.e., DF = 0, it is useless to use them, or their decay products, to measure DF's. Thus, in this experiment the only meaningful DF obtained was that for I^{131}. On March 11 it was $(4.0 \times 10^3)/120 = 33$, and on March 12 it was $(3.1 \times 10^3)/90 = 32$, both values being taken at 2.4-Mw power.

This is a surprisingly low value and indicates that some of the I^{131} in the reactor water is probably in the form of elemental iodine which is volatile, rather than in ionic form such as I^-, IO_3^-, or IO_4^-.

Previous work on the BORAX reactor[1,6] has shown a DF of 5×10^3 for Na^{24} at powers of 12-14 Mw, and higher for lower powers. There is no reason to suppose that the DF for nonvolatile species was any different during this test.

6. SUBSEQUENT FUEL INSPECTION

At the conclusion of the experiments outlined above, several months were devoted to the determination and location of the failed fuel elements.[5]

The only method which was considered to give conclusive results as to a defect in the sheath was to autoclave each fuel assembly individually. Each assembly was successively placed in an aluminum test container located in the center of one quadrant of the reactor. The test container was sealed, filled with clean deionized water, and the reactor was then operated at low power (10 kw) while air bubbled through the fuel assembly to carry out any fission gas which might be released. The effluent gas was monitored with a Geiger-Mueller counter.

After all fuel elements had been tested it was found that 22 assemblies had defects which could be detected. Each assembly was checked for a period of at least 30 min unless fission gases were detected sooner. In several elements, no signal was evident for as much as 20 min. Other elements indicated abnormal activities in less than 1 min.

The random location of defective elements within the reactor core is shown in Fig. 6-1.

Two of the elements which showed the highest activity of purge gas were sent to Argonne National Laboratory for metallurgical examination. The individual plates were cut from the fuel element in a remote-handling

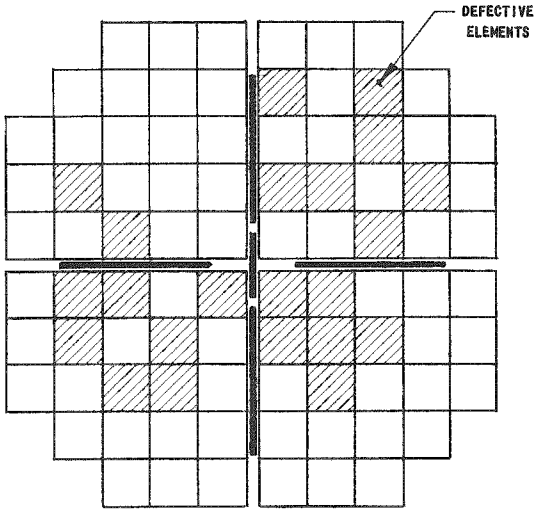
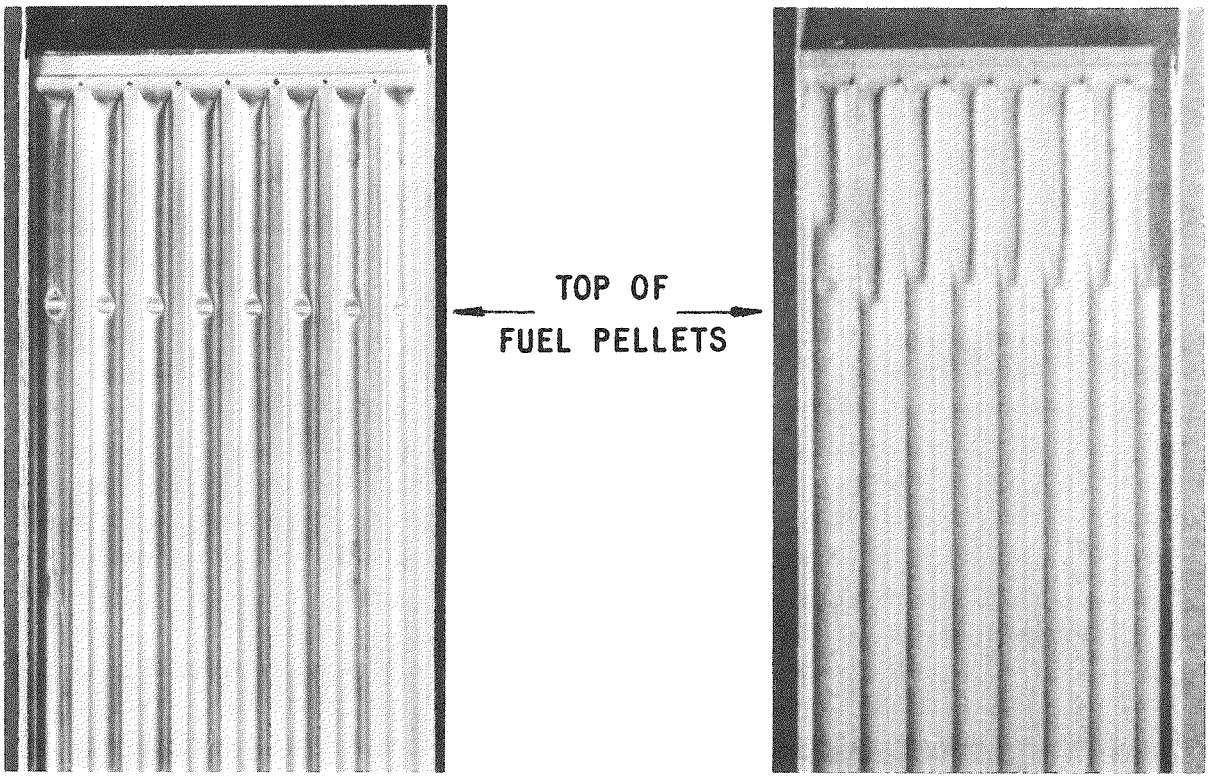


FIG. 6-1
DISTRIBUTION OF DEFECTIVE FUEL ELEMENTS IN CORE LATTICE

cave, and examination showed that in all plates the top of the individual pellet tubes had collapsed. Figure 6-2 shows a typical plate before irradiation and after removal from the reactor. It was found that several tubes had cracks through the cladding at the points indicated.

This report does not deal with the cause of this failure.[7] Suffice to say here that, of 69 elements in the reactor, 22 had failed to a greater or lesser degree.



BEFORE

AFTER

FIG. 6-2
TYPICAL APPEARANCE OF FUEL TUBES SHOWING COLLAPSE OF VOID REGION ABOVE FUEL PELLETS

7. DISCUSSION

7.1 Introduction

To establish the hazards due to radioactivity resulting from the operation of a boiling water reactor, such as BORAX IV, which contains defective ceramic-type fuel elements, three main questions must be answered:

(a) What is the source, nature, and magnitude of radioactivity released during operation?

(b) What is the nature and the level of the subsequent contamination ?

(i) of the reactor system?
(ii) of the steam system, including the turbine, condenser and piping leading to the stack?

In this problem only nuclides having half-lives longer than two or three hours need be considered.

(c) To what extent will the surrounding countryside be contaminated by long-lived fission products emitted from the reactor stack ?

In the following treatment answers to these questions will be attempted, with specific reference to the operation of BORAX IV.

The most significant results brought out by this experiment are:

(a) During operation of the reactor with the defective fuel, the main products which cause a high level of radioactivity in the system are the fission product gases and their decay products. Precursors such as radioiodines are presumably retained within the sheath (or the fuel) and there is no evidence that significant amounts of other fission products such as Mo^{99} which have no gaseous precursor are released through the defect.

(b) The results also indicate that there is a delay mechanism operating within the fuel such that shorter-lived gases are emitted to a lesser degree than the longer-lived ones.

Table 7-1 shows all decay chains which produce radioactive fission product gases. The chains are arranged in the order of increasing half-life of the fission gas. The data are taken from Reference 11; no chains involving stable gases have been included and several chains have been simplified in that branches of low yield involving short-lived activities have been neglected.

Examination of Table 7-1 shows that, if release of fission gas precursors through the defect is negligible, then the only chains which will contribute significantly to activity release and subsequent long-lived

Table 7-1

DECAY CHAINS INVOLVING RADIOACTIVE FISSION PRODUCT GASES

(* = Metastable)

Mass No.	Precursors	Fission Gas	Daughters	Yield, %
143		Xe (1s) →	Cs (short) → Ba (<0.5m) → Ld (18m) → Ce (33h) → Pr (13.7d) → Nd (stable)	6
94		Kr (1.4s) →	Rb (short) → Sr (2m) → Y (16.5m) → Zr (stable)	6.4
141		Xe (1.7s) →	Cs (short) → Ba (18m) → La (3.7h) → Ce (33d) → Pr (stable)	6
93		Kr (2.0s) →	Rb (short) → Sr (7m) → Y (10.5h) → Zr (1.1 x 10^6y)	6.5
92		Kr (3.0s) →	Rb (80s) → Sr (2.7h) → Y (3.6h) → Zr (stable)	6
91		Kr (10s) →	Rb (1.67m) → Sr (9.7h) → Y (58d) → Zr (stable) Rb (14m) → Sr (9.7h) → Y (58d) → Zr (stable)	5.8
140		Xe (16s) →	Cs (66s) → Ba (12.8d) → La (40,2h) → Ce (stable)	6.4
90		Kr (33s) →	Rb (2.7m) → Sr (28y) → Y (64.3h) → Zr (stable)	5.8
139	I (2.7s) →	Xe (41s) →	Cs (9.5m) → Ba (84m) → La (stable)	6.6
89	BR (4.5s) →	Kr (2.6m) →	Rb (15.4m) → Sr (51d) → Y (stable)	4.8
137	I (24.0s) →	Xe (3.9m) →	Cs (30y) $\xrightarrow{0.92}$ Ba* (2.57m) → Ba (stable) Cs (30y) $\xrightarrow{0.08}$ Ba (stable)	6.2
138	I (5.8s) →	Xe (17m) →	Cs (33m) → Ba (stable)	5.7
87	Se (17s) → Br (55s) →	Kr (78m) →	Rb (6 x 10^{10}y)	2.5
83	Se* (68s) → Br (2.4h) →	Kr* (114m) → Kr (stable)		0.5
88	Br (15.5s) →	Kr (2.8h) →	Rb (18m) → Sr (stable)	3.6
85	Se (40s) → Br (3.0m) →	Kr* (4.4h) $\xrightarrow{0.775}$ Rb (stable) Kr* (4.4h) $\xrightarrow{0.225}$ Kr (10.3y) → Rb (stable)		1.3
135	Te (<0.5m) → I (6.7h)	$\xrightarrow{0.3}$ Xe* (15.3m) → Xe (9.2h) → Cs (2.6x10^6y) → Ba (stable) $\xrightarrow{0.7}$ Xe (9.2h) → Cs (2.6x10^6y) → Ba (stable)		6.4
133	$\xrightarrow{0.72}$ Te* (63m) → Te (2m) → I (20.8h) $\xrightarrow{0.28}$ Te (2m) → I (20.8h)	I (20.8h) $\xrightarrow{0.02}$ Xe* (2.3d) → Xe (5.27d) → Cs (stable) I (20.8h) $\xrightarrow{0.98}$ Xe (5.27d) → Cs (stable)		6.6
131	$\xrightarrow{0.15}$ Te* (30h) → Te (24m) → I (8.05d) $\xrightarrow{0.85}$ Te (24m) → I (8.05d)	I (8.05d) $\xrightarrow{0.008}$ Xe* (12d) → Xe (stable) I (8.05d) $\xrightarrow{0.992}$ Xe (stable)		2.9

contamination are those which involve a gas of half-life shorter than 3.9 min, i.e., those above the chain of mass number 137 in the table. In all others, although the release rate of the gas itself may be high, the decay products which will produce contamination are either very short lived or so long lived they may be considered stable.

A more quantitative estimation will now be made. Results from defect tests of ceramic fuels in EBWR[4] indicate that if the quantity (R/Y), where R is the release rate of the gas in atom/sec and Y is the fission yield, is plotted against $\lambda^{-1/2}$, where λ is the decay constant, the data may be roughly correlated by a straight line passing through the origin.

In Fig. 5-1, the two experimental points obtained for Kr^{88} and Xe^{138} release are plotted. The correlation of these points by a straight line passing through the origin is not unreasonable. The release rates of other fission product gases will be calculated from this line; that is, knowing λ, the value of R/Y may be read from the plot, and hence R may be determined.

For purposes of calculation, the reactor system may be arbitrarily divided into five zones. As the gases are carried by the steam to the turbine and then by the exhaust gases to the stack, they will spend a certain time in each zone which will depend on the velocity of the carrier gas and the volume of the zone. The values listed below are believed to be quite typical of 2.4-Mw operation in BORAX-IV. Of course, for higher power operation all delay times will be proportionately smaller.

Zone	Location	Residence Time in Zone
1	Steam dome of reactor	30 sec
2	Piping from reactor to turbine, including dryer	30 sec
3	Turbine and condenser to air ejectors	30 sec
4	Piping from air ejectors to stack	$8\frac{1}{2}$ min
5	Atmosphere outside reactor	

Now, if R atom/sec for a given nuclide are released through the defects, then the number being released per second at the exit of each zone will be

$$R_1 = e^{-30\lambda} R \qquad R_3 = e^{-90\lambda} R$$
$$R_2 = e^{-60\lambda} R \qquad R_4 = e^{-600\lambda} R \tag{7.1}$$

The number of atoms decaying per second, i.e., the activity, in each zone will be

$$A_1 = R = R_1 = R(1 - e^{-30\lambda}) \qquad A_3 = R_2 = R_3 = R(e^{-60\lambda} - e^{-90\lambda})$$
$$A_2 = R_1 - R_2 = R(e^{-30\lambda} - e^{-60\lambda}) \qquad A_4 = R_5 - R_4 = R(e^{-90\lambda} - e^{-600\lambda}) \tag{7.2}$$

Using this model, the activities and resulting contamination in various parts of the system may be calculated.

7.2 Radioactivity during Steady-state Operation

Table 7-2 shows the release rates of the fission gases through the defect as calculated from Fig. 5-1. It also shows values for the various exponentials described above.

It will be assumed that the reactor has been operating for a sufficiently long time that steady-state conditions prevail in the steam and exhaust lines and that the release rate of fission gases from the defect is constant. This assumption is not justified in this experiment for the longer-lived fission gases; e.g., those with half-life longer than Kr^{87} (78 min), but for simplification of calculation it will be made. Thus activities and release rates of Kr^{87}, Kr^{83}, Kr^{88}, Xe^{138}, and Xe^{133} will be lower than those shown, and this calculation will represent the worst case. No account has been taken in the calculation for the removal of Xe^{135} by neutron capture.

Figure 7-1 shows a plot of the rate at which fission gases are arriving at or are being released from various locations in the system, calculated from Eq. 7.1. It is obvious that the very short-lived gases decay completely in the steam dome, and the majority of the activity in the remainder of the system will be due to fission gases having half-lives from 10 sec to 1 hr, and to the decay products from these gases.

A check may now be made to see whether the calculated data of Table 7-2 and Fig. 7-1 are in accord with observed results of this test.

Table 4-4 shows the activities of several nuclides found in the reactor steam at sampling time. For instance, the Cs^{137} activity was about 20 d/(min)(ml) or 0.33 d/(sec)(ml). Thus at sampling there were $0.33/7.33 \times 10^{-10} = 4.5 \times 10^{8}$ atom/cc or roughly 4.5×10^{8} atom/gm of condensed steam present. It will be assumed that the Cs^{137} resulted from the decay of Xe^{137} which was in the steam at the time the samples were taken. Thus 4.5×10^{8} atom/gm of Xe^{137} were present in the steam. At a steam flow rate of 958 gm/sec, the rate at which Xe^{137} atoms were passing the sampling point was $4.5 \times 10^{8} \times 958 - 4.3 \times 10^{11}$ atoms/sec. Table 7-3 shows the results of this type of calculation applied to the Ba^{140}, Sr^{90}, and Cs^{137} activities found in steam samples. Also shown in this table are the rates (calculated from the data of Table 7-2) at which the fission gases are entering and leaving the steam line.

Evidently the calculations outlined in this and the preceding section are conservative in that they predict higher release of the fission gases than actually occur. Also the ratio of the calculated rate to the observed rate appears to increase as the half-life of the fission gas decreases.

Table 7-2

CALCULATED RELEASE RATES OF FISSION GASES

Chain	Nuclide	Half life	λ, sec^{-1}	$\lambda^{-1/2}$ sec	Release Rate to Water		A[a]	B[a]	C[a]	D[a]	1 - A	A - B	B - C	C - D
					x 10^{11} atom/sec	curie/sec								
143	Xe	1 sec	0.693	1.20	1.07	2.0	neg	neg	neg	neg	1	neg	neg	neg
94	Kr	1.4 sec	0.495	1.42	1.34	1.8	neg	neg	neg	neg	1	neg	neg	neg
141	Xe	1.7 sec	0.407	1.57	1.39	1.5	neg	neg	neg	neg	1	neg	neg	neg
93	Kr	2.0 sec	0.346	1.70	1.64	1.5	neg	neg	neg	neg	1	neg	neg	neg
92	Kr	3.0 sec	0.231	2.08	1.85	1.2	0.001	neg	neg	neg	0.999	0.001	neg	neg
91	Kr	10 sec	6.93×10^{-2}	3.81	3.27	0.61	0.125	0.015	neg	neg	0.875	0.110	0.015	neg
140	Xe	16 sec	4.33×10^{-2}	4.80	4.54	0.53	0.27	0.075	0.020	neg	0.730	0.195	0.055	0.020
90	Kr	33 sec	2.10×10^{-2}	6.90	5.91	0.34	0.534	0.284	0.152	3.4×10^{-6}	0.466	0.250	0.132	0.152
139	Xe	41 sec	1.69×10^{-2}	7.69	7.52	0.34	0.603	0.362	0.218	neg	0.397	0.241	0.144	0.218
89	Kr	2.6 min	4.45×10^{-3}	15.0	10.7	0.13	0.875	0.765	0.670	0.069	0.125	0.110	0.095	0.601
137	Xe	3.9 min	2.96×10^{-3}	18.4	16.8	0.14	0.915	0.838	0.767	0.17	0.085	0.077	0.071	0.597
138	Xe	17 min	6.80×10^{-5}	38.3	35.9	0.07	0.98	0.96	0.94	0.665	0.02	0.02	0.02	0.275
87	Kr	78 min	1.48×15^{-4}	82.3	30.0	0.01	0.996	0.992	0.987	0.915	0.004	0.004	0.005	0.072
83	Kr	114 min	1.01×10^{-4}	99.5	7.35	2×10^{-4}								
88	Kr	2.78 hr	6.93×10^{-5}	120	61.9	0.01	0.998	0.996	0.994	0.96	0.002	0.002	0.002	0.034
85	Kr	4.4 hr	4.38×10^{-5}	151	29.1	3.5×10^{-3}	0.999	0.998	0.996	0.974	0.001	0.001	0.002	0.026
135	Xe	9.2 hr	2.09×10^{-5}	218	322	1.2×10^{-3}	1.0	1.0	1.0	1.0	neg	neg	neg	neg
133	Xe	5.27 d	1.52×10^{-6}	812	780	3.2×10	1.0	1.0	1.0	1.0	neg	neg	neg	neg

[a] $A = e^{-30\lambda}$ $\quad$ $C = e^{-90\lambda}$

$B = e^{-60\lambda}$ $\quad$ $D = e^{-600\lambda}$

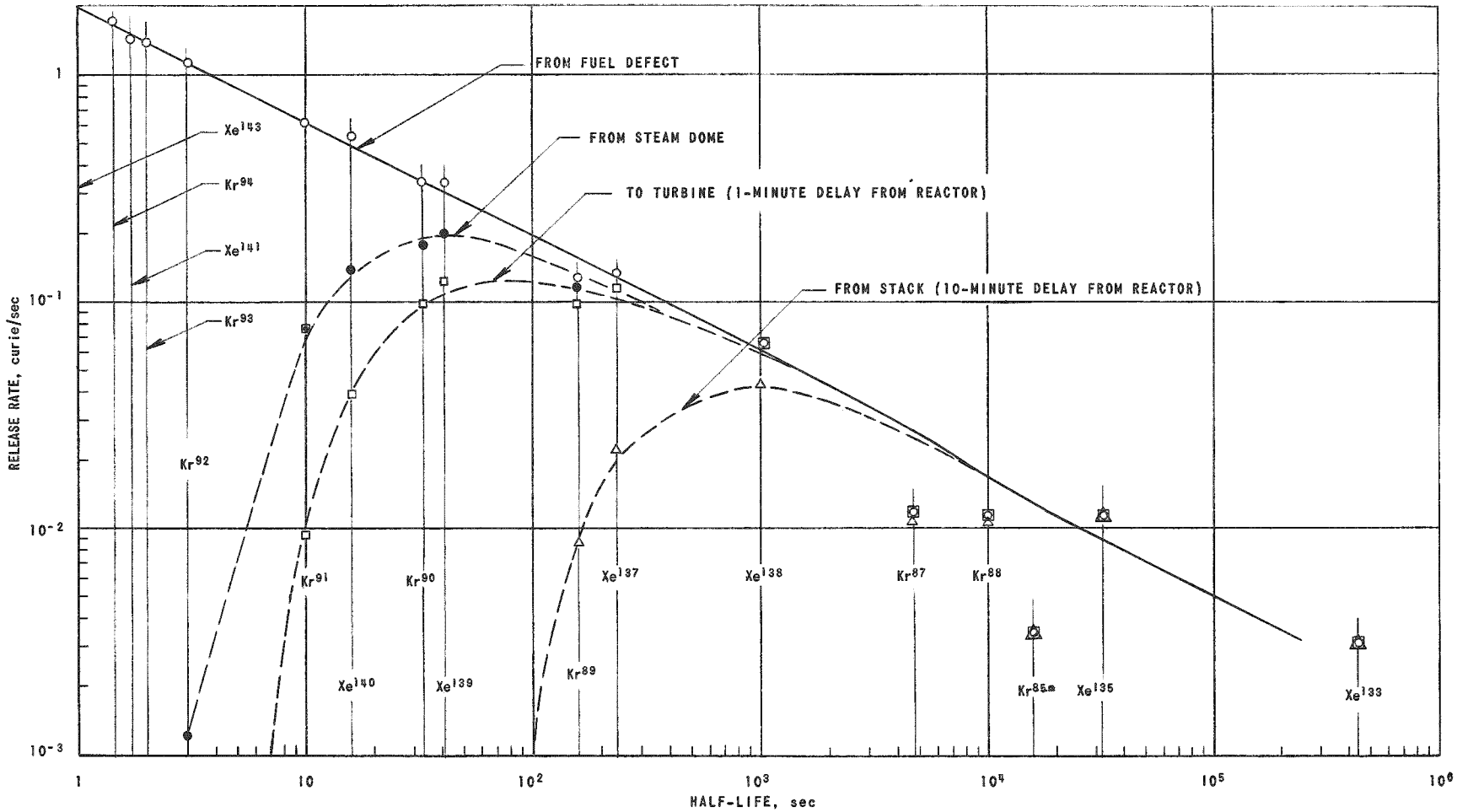


FIG. 7-1
RELEASE OF FISSION GASES AT VARIOUS LOCATIONS IN BORAX-IV AT 2.4 Mw

Table 7-3

COMPARISON OF CALCULATED AND MEASURED RATES OF SHORT-LIVED FISSION GAS RELEASE TO AND FROM STEAM PIPE

Nuclide	Half-life	Measured Rate at Sampling Point (atom/sec)	Calculated Rate (atom/sec) Into Steam Pipe	Calculated Rate (atom/sec) From Steam Pipe
Xe^{140}	16 sec	3.1×10^{9}	1.2×10^{11}	3.4×10^{10}
Kr^{90}	33 sec	2×10^{11}	3.2×10^{11}	1.7×10^{11}
Kr^{89}	2.6 min	2×10^{11}	9.4×10^{11}	8.2×10^{11}
Xe^{137}	3.9 min	4.4×10^{11}	1.5×10^{12}	1.4×10^{12}

This would indicate that for the shorter-lived gases the rate of release falls even faster than would be indicated by a plot of R/Y versus $\lambda^{-1/2}$. Another explanation would be that the delay times in the steam dome and steam pipe have been underestimated.

However, since the proposed mechanism leads to conservative results it will be used in further calculations.

Figure 7-2 shows a plot of the activity, in curies, due to fission gases in various parts of the system as calculated from Eq. 7.2. Again, as expected, the farther in time one gets from the reactor vessel, the longer lived the predominant fission gas activity becomes. Note that during steady operation, there will be at least 12 curies of fission gases alone in the turbine and condenser. Short-lived daughters will bring this up to well over 30 curies of activity.

These figures illustrate why Cs^{138} was such a predominant activity during reactor operation. Figure 7-1 shows that Xe^{138} was released from the stack at a significantly higher rate than any other gas and hence Cs^{138} will be the predominant short-lived activity outside the reactor. In the rest of the system it will be overshadowed by other gases and their daughters during actual operation.

On shutdown, however, the shorter-lived nuclides will decay quite rapidly, and the longer-lived nuclides have no significant daughters. Thus one-half hour or so after shutdown, Cs^{138} will be the predominant activity, especially in the turbine and condenser and in the piping beyond the condenser.

In Fig. 7-2, the points for N^{16} were calculated using data obtained by M. Theys from EBWR operation.[8] At 2.4 Mw the N^{16} production rate may be calculated to be 7×10^{11} atoms/sec. If 10% of this goes

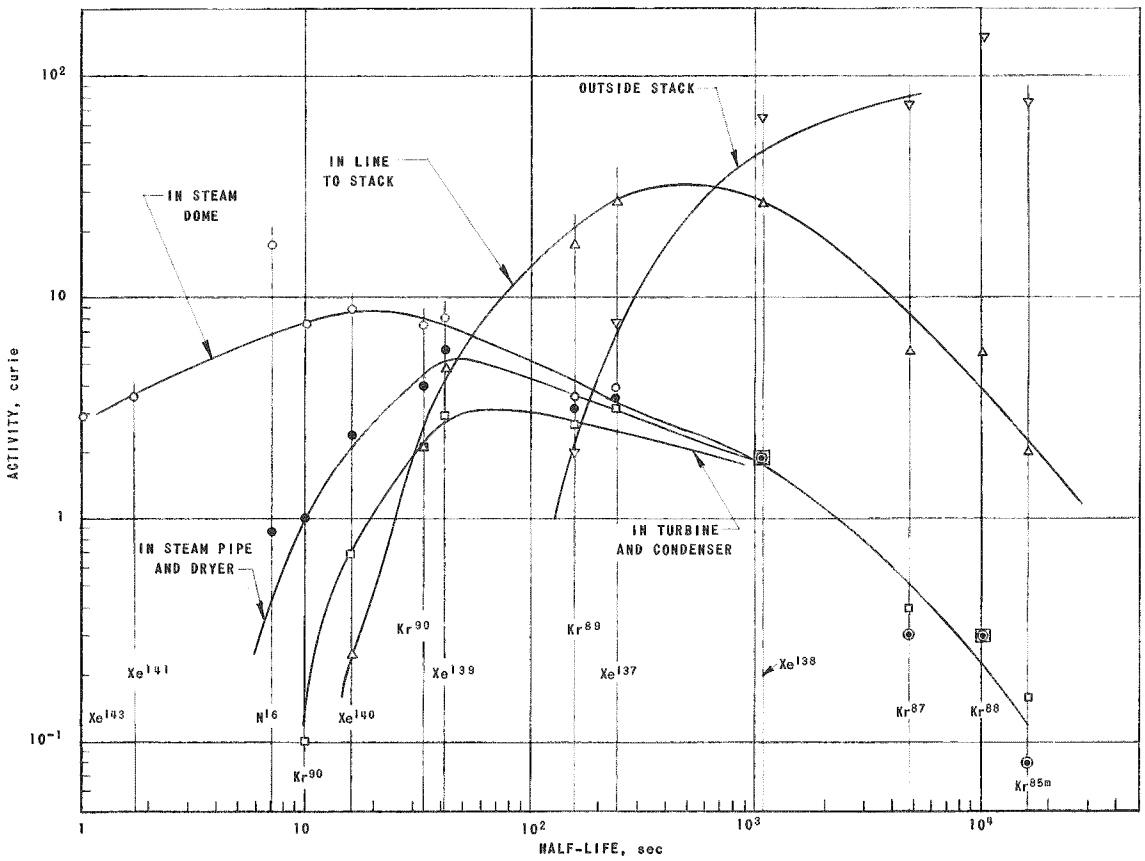


FIG. 7-2
DISTRIBUTION OF FISSION GASES IN BORAX-IV AT 2.4 Mw

into the steam then the production rate in the steam dome is 7 x 10^{10} atom/sec. Figure 7-2 illustrates clearly that in the steam dome N^{16} is still a predominant activity, but once the steam leaves the reactor vessel the N^{16} activity is overshadowed by that of fission gases, and when the steam reaches the turbine the N^{16} is a negligible contributor to the total activity.

7.3 Long-lived Contamination

In making the calculations regarding the extent of long-lived contamination it will be assumed that all contamination results from long-lived daughters of fission gases. Contamination from other sources will be discussed in the appropriate sections.

It will be assumed that the reactor has operated steadily at 2.4 Mw for eight hours prior to shutdown and that the release rate of fission gas, R, to the water is constant. In any one of the zones outlined above, the number of fission gas atoms decaying per second will be given by Eq. (7.2). Knowing the decay chains, the number of curies of long lived decay product at any given time after reactor shutdown may be calculated. This calculation is outlined in Appendix I and the results are presented in Table 7-4.

Table 7-4

CALCULATED LONG-LIVED CONTAMINATION RESULTING FROM 8 HR OPERATION AT 2.4 Mw

(All Activities in Millicuries)

Nuclide	Half-life	1 Day after Shutdown					10 Days after Shutdown				
		Zone 1	Zone 2	Zone 3	Zone 4	Zone 5	Zone 1	Zone 2	Zone 3	Zone 4	Zone 5
Ce^{143}	33 hr	300	neg	neg	neg	neg	3	neg	neg	neg	neg
Pr^{143}	13.7 d	20	neg	neg	neg	neg	30	neg	neg	neg	neg
Ce^{141}	33 d	25	neg	neg	neg	neg	20	neg	neg	neg	neg
Sr^{91}	9.7 hr	0.790	100	10	neg	neg	neg	neg	neg	neg	neg
Y^{91}	58 d	25	3	0.5	neg	neg	27	3	0.5	neg	neg
Ba^{140}	12.8 d	150	41	12	4	neg	95	25	7	3	neg
La^{140}	40.2 hr	53	14	4	2	neg	110	29	8	3	neg
Sr^{90}	28 yr	0.17	0.09	0.05	0.06	1.2×10^{-6}	0.17	0.09	0.05	0.06	1.2×10^{-6}
Y^{90}	64.3 hr	0.04	0.02	0.01	0.01	neg	0.15	0.08	0.04	0.05	neg
Sr^{89}	51 d	16	14	12	78	9	14	12	10	66	8
Cs^{137}	30 yr	0.005	0.04	0.035	0.3	0.085	0.005	0.04	0.035	0.3	0.085
Total Millicuries		1380	190	40	85	9.3	300	70	25	72	8.3

Zone 1 - Reactor Steam Dome

Zone 2 - Steam Pipe and Dryer

Zone 3 - Turbine and Condenser

Zone 4 - Piping to Stack

Zone 5 - Outside Stack

One assumption inherent in the above calculation is that a decay atom from a fission gas remains within the zone in which it is born. This may not be so in that some may be swept by the steam into another zone, and perhaps ultimately back to the reactor via the condenser hotwell and feedwater pumps. However, this calculation will give the worst case results assuming all decay products remain "in situ."

For the long-lived fission gases the assumption that a steady release rate will be achieved in only eight hours operation is not justified. Fortunately these long-lived fission gases do not lead to any serious contamination. Table 7-1 shows that decay products from all gases of half-life longer than 17 min are all short-lived, leading to either stable or very long-lived isotopes.

Examination of Table 7-1 shows that only chains of mass numbers 143, 141, 140, 90, 89, and 137 need be considered, and Table 7-4 shows the results for each zone calculated for one and ten days after reactor shutdown.

Each zone will now be discussed in detail.

7.3.1 Zone 1 - Reactor Vessel

Contamination in the reactor vessel will come from three sources:

(a) The first source will, of course, be from the decay of fission gases outlined above. It is interesting to note that the majority of the long-lived activity remains within the reactor vessel. Also, it is interesting to note how low this level is. Fourteen curies of activity in the reactor vessel is negligible compared with activities of activated corrosion products which are there already.

It must be remembered that if any of the decay products are not deposited in situ they may deposit further down the system on the steam piping or in the turbine.

(b) The second source of contamination is from fission gas decay products which have been swept from piping, turbine, etc., and returned to the reactor via the feedwater. It is known (Table 4-4) that the feedwater contained Cs^{137}, Cs^{138}, Ba^{140}, and Sr^{89}.

The net result of this would be that the contamination in Zones 2 to 4 may be lower than indicated but the activity in Zone 1 will not be increased greatly.

(c) The third source of contamination is the presence of nonvolatile components which have been released from the fuel. During this test these components evidently contributed to the activity of the reactor water to a minor extent. This has also been found to be true for UO_2 ceramic fuels,[9,10] with the exception that they seem to release large quantities of radioiodines, comparable to the amounts of fission gas released. This does not appear to have been true during this test. ThO_2 may actually release radioiodines to a lesser extent than does UO_2 or the effect may be due to their absorption on Al or Al_2O_3 surfaces. It must be remembered that all UO_2 tests reported have been conducted in steel systems with no aluminum present.

7.3.2 Zone 2 - Steam Pipe; Zone 3 - Turbine and Condenser; and Zone 4 - Piping to Stack

Table 7-4 shows that the main source of contamination in these zones comes from Ba^{140}-La^{140}, Sr^{90}-Y^{90}, Sr^{89}, and Cs^{137}. It is significant that these nuclides were the only ones found in steam and condensate samples. Also γ-ray pulse-height analyses of the samples from the turbine (Section 4.4) showed only Ba^{140}-La^{140} and Cs^{137}. (Sr^{89} and Sr^{90} have no γ rays.) These results indicate that contamination from such nuclides as Ce^{143}, Pr^{143}, Ce^{141}, or Y^{91} is not serious in this part of the system.

A rough check can be made to compare the calculated data of Table 7-4 with the observed contamination on the metal coupons in the turbine (Table 4-6). Below are shown the ratios of the Ba^{140} and Cs^{137} activities, calculated for one day after shutdown. The first three numbers, for Zones 2, 3, and 4, are obtained from Table 7-4. The last number, that for the metal coupons, is taken from Table 4-6.

	Ratio $\frac{\text{Activity } Ba^{140}}{\text{Activity } Cs^{137}}$
Zone 2 - Steam Pipe	~ 500
Zone 3 - Turbine and Condenser	170
Zone 4 - Piping to Stack	7
Metal Coupons in Turbine	10 to 13

The measured ratio appears to agree better with that calculated for the piping to the stack. If, however, the delay time between the reactor and the turbine were actually longer than that assumed, then the calculated results would fall in line better. The discrepancy would also be accounted for if the release rate for very short-lived gases fell more steeply than indicated by the R/Y versus $\lambda^{-1/2}$ plot. However, here again the proposed mechanism leads to conservative results.

If the surface area in the turbine and condenser is arbitrarily assumed to be 10^4 ft^2, then from Table 7-2 the Cs^{137} activity in the turbine should be about 0.007 $\mu c/ft^2$. Table 4-6 showed the Cs^{137} on the mild steel coupons was about 0.03 $\mu c/ft^2$. Considering all the uncertainties, the agreement is at least fair.

Examination of Table 7-4 reveals a few more interesting facts. For instance, almost one-half of all the Sr^{90} produced remains in the steam dome, the remainder being found mainly in the steam line, with some in the turbine and piping to the stack. Virtually none escapes from the stack. Sr^{89}, however, deposits mainly in the line to the stack.

Thus, it must be remembered that these two nuclides, which are the most harmful from radiological health standpoint, will be present on the walls of the steam pipe, in the turbine and condenser and in the line to the stack. Due cognizance of this fact must be taken when these parts are opened up after reactor shutdown.

Contamination in these zones might also arise from the presence of nonvolatile (or slightly volatile, such as radioiodine) nuclides present in the reactor water, which are physically entrained in the steam. This test and previous experiments, however, indicate that the DF between steam and water is greater than 200 and more probably in the order of 10^3 or 10^4. Thus buildup of long-lived contamination from this source will not be important.

7.3.3 Contamination Released from Stack

During this test very little long-lived contamination was released from the stack. Of course, large amounts of short-lived activity, such as from Xe^{138} and Cs^{138} will be released, but in a few hours these will have decayed to negligible levels.

Large amounts of long-lived fission gas will also be released (Figs 7-1 and 7-2) but these are not considered a potential health hazard since they decay to stable or very long-lived nuclides. They should be dispersed into the atmosphere and diluted very rapidly.

The most important contaminants from a health point of view are Sr^{90} and Sr^{89}. It is obvious that the ten-minute delay from the reactor to the stack is enough to make the resulting outside contamination by Sr^{90} negligible. Some Sr^{89} will build up outside. The amount that results from 8-hour operation, 8 millicuries, will be spread over a large area and should not pose a large problem.

8. SUMMARY

The first important general conclusion from the results of this test is that it is perfectly feasible to operate a boiling water reactor fueled with ceramic fuel with many defects in the fuel cladding such as were present during this test.

Although radiation levels may be high during operation, it would appear that protracted operation would be quite feasible without serious contamination to equipment or to the surrounding countryside providing two criteria were met:

a. The system should be free from leaks.
b. The delay time from the reactor vessel to the exit from the stack should be longer than one hour.

With such a delay time the amounts of Sr^{89}, Sr^{90}, or Cs^{137} released from the stack would be negligibly small, even for a large number of defected elements. If the delay time were of the order of 24 hr, then the only significant activities released from the stack would be due to Xe^{135} and Xe^{133}.

The second important conclusion is that a means exists of estimating the activity release and the resulting contamination from operating reactor with defective ceramic fuel.

First of all, the only fission products which are released in significant quantities are fission gases. If a release rate for one gaseous nuclide can be measured or estimated, then the release rates of all other fission gases may be calculated. This calculation will lead to conservative results in that estimated release rates for very short-lived fission gases appear to be larger than they actually are. Then, if the reactor, steam, turbine and condenser, and off-gas systems are divided into a number of zones, and an average time that a gas atom will spend therein, assigned to each zone, then the activities throughout the system at steady operation may be estimated and the long-lived contamination resulting from a known period of operation may be calculated, both for the reactor system and for the surrounding countryside..

APPENDIX

CALCULATION OF LONG-LIVED ACTIVITIES IN VARIOUS PARTS OF REACTOR STEAM SYSTEM

In Section 7.1 it was shown that the decay rate R' of a fission product gas in any zone will be given by:

$$R' = R(e^{-\lambda t_1} - e^{-\lambda t_2}) \quad ,$$

where

R = Release rate of fission gas from defect (atoms/sec)

λ = Decay constant of that gas (sec^{-1})

t_1, t_2 = Delay times from the defect to the zone entrance and to the exit respectively.

It will be assumed that the fission gas A is the precursor of a chain:

$$A \xrightarrow{\lambda_A} B \xrightarrow{\lambda_B} C \xrightarrow{\lambda_C} \text{etc.} \quad ,$$

where the λ's are the decay constants of the respective nuclides.

The activities of various nuclides may now be calculated at various times while the reactor is operating and after it has been shut down.

A. Reactor Operating

It will be assumed that the reactor has been operating at steady power for a time T hours prior to shutdown.

In any zone:

$$\frac{dA}{dt} = R' - \lambda_A A$$

$$\frac{dB}{dt} = \lambda_A A - \lambda_B B$$

$$\frac{dC}{dt} = \lambda_B B - \lambda_A A \quad \ldots\ldots$$

In these equations, A, B, C, etc. represent the number of atoms of each nuclide in that zone. Implicit in these equations is the assumption that any nuclide formed in a zone decays in that zone and that none enter or leave by any other means.

These equations may be solved by well-known methods, using the boundary condition $t = 0$, $A = B = C = \cdots\cdots = 0$. Fortunately, in all chains of interest, the calculation is simplified by the fact that the half-lives of the first few members are short compared to the length of time the reactor has operated and hence steady-state conditions prevail.

Thus the number of atoms of each nuclide at time T, the time of reactor shutdown, can be calculated. These are designated by A^0, B^0, $C^0, \cdots\cdots$.

B. Reactor Shutdown

When the reactor is shut down a similar set of equations may be written, except that $R' = 0$:

$$\frac{dA}{dt} = -A\lambda_A$$

$$\frac{dB}{dt} = A\lambda_A - B\lambda_B$$

$$\frac{dC}{dt} = B\lambda_B - C\lambda_C \quad \cdots\cdots$$

Again, these equations may be solved by well-known methods[12] using the boundary conditions, $t = 0$, $A = A^0$, $B = B^0$, $C = C^0, \cdots\cdots$.

Thus the number of atoms, and hence the activity, of A, B, C, $\cdots$, may be calculated at any time after reactor shutdown.

These calculations may appear rather tedious, but they are shortened considerably in most chains by the fortuitous circumstance that the first few members of the chain are very short lived and then there is an abrupt transition from short-lived to long-lived nuclides.

REFERENCES

1. W. H. Zinn, et al., "Operational Experience with the BORAX Power Plant," Nuclear Science and Engineering, Vol. 1, No. 5 (October, 1956).

2. J. H. Handwerk and R. A. Noland, "Oxide Fuel Elements for BORAX IV," Paper presented at the Technical Briefing Session, held at Argonne National Laboratory, May 27-28, 1957, TID-7535, p. 140.

3. B. S. Maxon, O. A. Schulze, and J. A. Thie, "Reactivity Transients and Steady-state Operation of a Thoria-Urania-fueled Direct-cycle Light Water-Boiling Reactor (BORAX IV)," ANL-5733 (February, 1959).

4. R. F. S. Robertson, "Tests of Defected Thoria-Urania Fuel Specimens in EBWR," ANL-6022 (To be published).

5. G. K. Whitham, W. R. Wallin, and E. Graham, "Locating Defective Fuel Elements in BORAX-IV." Paper presented at the American Nuclear Society Meeting, held in Gatlinburg, Tennessee, June 15-17, 1959.

6. A. J. Shor, et al., "Radioactive Carry-over from BORAX-III and Test Systems," Nuclear Science and Engineering, Vol. 2, No. 2 (April, 1957).

7. C. F. Reinke, "Examination of BORAX-IV Defective Fuel Plates" (To be published).

8. M. H. Theys and R. L. Mittl, "Experimental Study of N^{16} Concentrations in Water and Steam of EBWR Pressure Vessel." Paper presented at the American Nuclear Society Meeting held at Gatlinburg, Tennessee, June 15-17, 1959.

9. J. D. Eichenberg, et al., "Effects of Irradiation on Bulk UO_2," WAPD-183 (October, 1957).

10. J. A. L. Robertson, et al., "Behavior of Uranium Oxide as a Reactor Fuel," Paper No. 193 presented at the Second International Conference on the Peaceful Uses of Atomic Energy held in Geneva, Switzerland, September, 1958.

11. "Reactor Physics Constants," ANL-5800.

12. G. Friedlander and J. W. Kennedy, "Introduction to Radiochemistry," John Wiley and Sons, Inc., New York, (1949), Ch. V.

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