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DECONTAMINATION OF THE HOMOGENEOUS  
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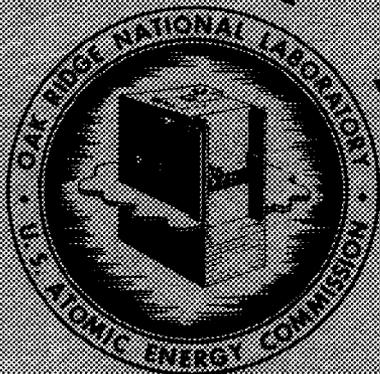
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CHEMICAL TECHNOLOGY DIVISION  
Chemical Development Section

DECONTAMINATION OF THE  
HOMOGENEOUS REACTOR EXPERIMENT

D. O. Campbell

DATE ISSUED:

MAY 22 1956

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## 1.0 ABSTRACT

After shutdown the Homogeneous Reactor Experiment system was decontaminated in about one month, without descaling, from an activity level of 1000 r/hr to 5-200 r/hr. This was sufficient to permit dismantling. The treatment consisted in washing twice with each of the reagents 5% nitric acid, 35% nitric acid, and 10% sodium hydroxide--1.5% sodium tartrate--1.5% hydrogen peroxide and numerous times with water. Overall decontamination factors were 22-25, including decay. Decontamination factors with a single reagent were between 1 and 2.25. More than 2000 curies each of cerium and zirconium, more than 1000 curies each of barium, strontium, and lanthanum, and large amounts of niobium, ruthenium, and iodine were removed. The significant contaminants remaining were niobium and zirconium. These could have been removed only by descaling the system, which laboratory experiments indicated would have given a further decontamination factor of about 100.

## 2.0 INTRODUCTION

Shortly after shutdown, the radiation readings in the Homogeneous Reactor Experiment (HRE) cells were of the order of 1000 r/hr. The objective of the work reported here was to decontaminate the system to an activity level sufficiently low to permit dismantling it within one month.

Previous decontamination experience at ORNL had been concerned largely with type 347 stainless steel contaminated at temperatures below the boiling point by uranyl nitrate--nitric acid solutions of fission products that had decayed for several months (Purex type systems). Zirconium and niobium were the most important contaminants. The HRE system was also of type 347 stainless steel, but the fuel was a uranyl sulfate--sulfuric acid solution and the system was operated at a temperature near 250°C. The fission products had decayed for only a few days, and short-lived isotopes were present. Corrosion is severe in such a system, and under HRE operating conditions a protective oxide film forms on type 347

stainless steel. It was necessary that the decontamination be accomplished without descaling to be certain that corrosion by the decontamination solutions would not obscure subsequent studies on corrosion caused by the reactor fuel solution.

The author would like to express his appreciation to all persons responsible for obtaining the information reported here, in particular, J. J. Hairston and his group at the HRE site, who did most of the work on which the results are based; members of the Analytical Chemistry Division, especially E. I. Wyatt and his group who performed the many difficult radiochemical analyses and S. A. Reynolds for his aid in interpreting the data; and the Oracle staff of the Mathematics Panel, particularly S. G. Campbell, who coded the information for calculation of the amounts of fission products produced by the HRE.

### 3.0 DECONTAMINATION PROCEDURE

On the basis of laboratory experiments (Sec. 8.0) and the previous decontamination experience with Purex type systems, the following decontamination procedure for the HRE was suggested:

The fuel solution should be thoroughly washed from the system with water, and the system should then be treated alternately with 35% nitric acid for at least 4 hr and 10% sodium hydroxide--1.5% sodium tartrate--1.5% hydrogen peroxide for a longer time, preferably overnight. Water washes should be used after each reagent treatment. The volume of each solution should be at least 200 liters, the volume of the fuel when the reactor was operating. It is essential that all contaminated surfaces be contacted with the solutions.

The HRE equipment was in four separate cells: the heavy water cell, with which the work was not concerned; the reactor cell, containing the core and reflector vessels; the heat exchanger cell, containing the heat exchanger and the fuel circulating pump; and the fuel solution cell, containing the

dump tank, the pump that brought fuel from the dump tank to the high-pressure system, the fuel make-up equipment, the gas system, and the rest of the associated equipment. The inner dump tanks were at the bottoms of the cells and were not shielded from the cell interiors.

The decontaminating reagents were introduced into the dump tank, from which they were pumped into the circulating fuel system, being backwashed through the system filter. During operation of the reactor, this filter presumably had collected any insoluble fission products and detached corrosion scale, which carries some fission products and was probably one of the most radioactive parts of the system. It is unfortunate that the decontaminating solutions had to contact a highly radioactive part of the system first and therefore probably carried some fission products from the filter through the rest of the system. This would be expected to decrease the efficiency of the decontaminating solution for the rest of the system. All solutions were introduced at or near room temperature, but the circulating pump heated them to about 70°C. After the reagent had circulated for the desired time, it was drained. Since a heel of about 10 liters remained, the subsequent water wash was really a dilute decontaminating reagent. A 25-ml sample of each decontaminating reagent was removed just before the solution was dumped, and the radiation reading at contact was determined. This reading is a rough measure of the gamma-emitting fission product removed by the reagent. Part of this sample was later analyzed for individual fission products.

After the solution had been drained, the radiation level in each cell was measured with a probe dropped through a hole in the shielding plug at the top of the cell. The probe was 9.5 ft down in the reactor and heat exchanger cells and 19.5 ft down in the fuel solution cell. The readings therefore depended largely on the radioactivity of the equipment closest to the probe, but the readings for any one cell at different times may be compared with

some significance.

The reactor had operated for a total of 580 Mw-hr and had produced about 25 g of fission products (Sec. 9.2). It operated for most of its power in December 1953 and January 1954. It was shut down at 9 AM, March 3, 1954, after 40 hr continuous operation at 400 kw. The activity levels were reduced by a factor of about 10 as the reactor was made subcritical. The fuel solution cell activity level decreased by a factor of 3 because its level was more dependent on fission product activity than on prompt activity (i.e., that emitted at the time of fission or neutron capture). From the readings taken after the fuel was drained, it is obvious that a large part of the gamma-emitting fission products remained in the system and did not follow the fuel solution.

The reactor system was washed with water nine times between shutdown and the beginning of the decontamination. The rinse water was condensate from distillation of the fuel solution, and volatile fission products therefore may have been reintroduced into the system with each wash. The last rinse solution was sampled for radiochemical analysis. The decrease of activity during rinsing, by a factor of 7-20, was the result of both decay and, especially for the first rinse, decontamination.

After the rinse, the fuel dump tanks were isolated from the rest of the system by freezing water in the drain line, and decontamination was started on March 18. Two hundred liters of reagent was used in each case until near the end, when the approximately 500-liter system was completely filled with four of the solutions (Nos. 14, 15, 20). The temperature was increased to about 100°C for some of the water washes. Decontamination was concluded on April 14, with dumping of the final rinse solution at 4 PM.

When the top plugs of the cell shielding had been removed, after the decontamination, the activity over the cells, at floor level on top of the shield, varied from a maximum of 5 r/hr, in

a fairly well defined beam apparently coming from the part of the bottom of the fuel cell that was not shielded by the cell equipment, to a few hundred milliroentgens per hour elsewhere. Most of the dismantling was done in radiation levels that varied from a few to 200 mr/hr. The equipment was cut apart with an air-driven hacksaw or a cutting torch on the end of a long pipe, the resulting pieces being removed with the overhead crane. The decontamination was sufficient to permit this sort of destructive operation, but the more rigorous requirements of equipment repair, replacement, or modification probably could not have been met without further decontamination. The dismantling was completed several weeks ahead of schedule. Radiation levels of the system components, after the dismantling, are given in Table 15.

#### 4.0 INDIVIDUAL REAGENT AND OVERALL DECONTAMINATION FACTORS

It was estimated that an overall decontamination factor of about 10 should be obtained by the recommended procedure. The procedure actually carried out resulted in overall decontamination factors, including about six weeks' decay, of 22-25 for the three cells concerned (Fig. 1, Tables 1 and 14). With descaling (Sec. 5.0), a decontamination factor of the order of 2000 could probably have been obtained, provided that all parts of the system could have been properly contacted by the reagents.

Most individual reagent decontamination factors were slightly greater than 1; a few were less, indicating that activity was transferred into the cell under some conditions. Cell activity measurements were subject to considerable error, which could account for some of the variation. In general, larger decontamination factors were obtained when a solution was used for the first time or when some other condition was changed from what it had been for previous treatments. Thus, with the first 35% nitric

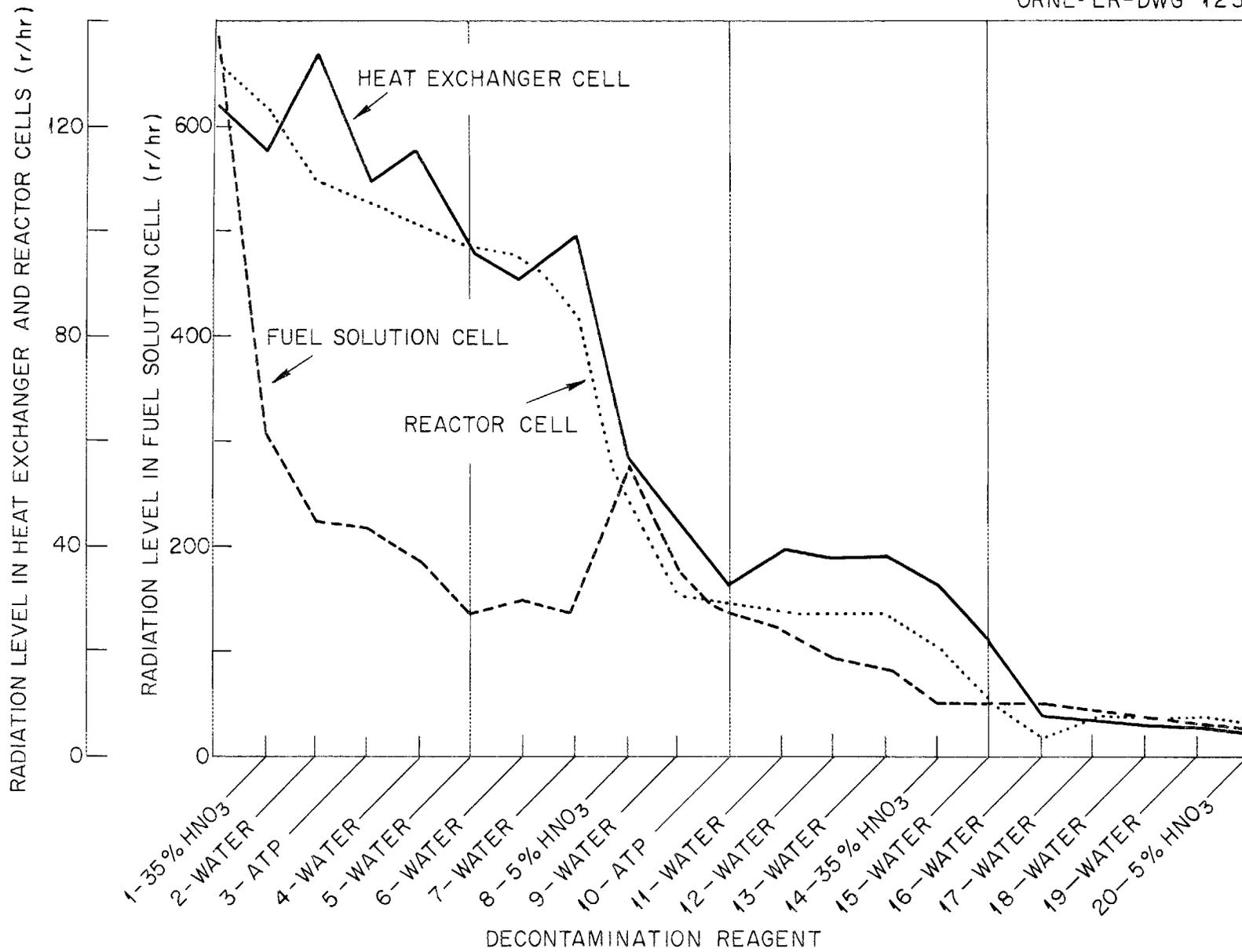


Fig. 1. Radiation Levels of HRE Cells during Decontamination.

Table 1. Radiation Levels of HRE Cells during Decontamination

	Date	Time	Radiation Level (r/hr)		
			Fuel	Reactor	Heat
			Solution Cell	Cell	Exchanger Cell
While operating at 400 kw	3/3/54	7:30 AM	47,000	33,900	27,300
After shutdown at 9:00 AM and draining of fuel	3/3	12:00 N	5,300	1,050	1,285
After 1st water rinse	3/4	3:55 PM	2,140	575	536
After 9th water rinse	3/16	4:00 PM	710	150	140

Reagent	Reagent No.	Length of Treatment (hr)	Date Reagent Removed	Reading of 25 ml of Reagent just before Dumping (r/hr)	Fuel Solution Cell		Reactor Cell		Heat Exchanger Cell	
					(r/hr)	D.F.	(r/hr)	D.F.	(r/hr)	D.F.
---	---	---	---	---	690	---	132	---	125	---
35% HNO <sub>3</sub>	1	2	3/18	60	306	2.25	123	1.27	115	1.09
Water	2	4	3/19	3.0	224	1.35	109	1.13	137	0.87
ATP <sup>a</sup>	3	70	3/23	2.6	213	1.04	104	1.04	109	1.21
Water	4	3	3/24	1.6	192	1.14	98	1.04	115	0.94
Water	5	6	3/25	2.0	137	1.37	93	1.04	93	1.24
Water	6	5	3/26	1.5	148	0.94	96	1.01	91	1.05
Water	7	50	3/29	0.6	143	1.03	85	1.19	99	0.87
5% HNO <sub>3</sub>	8	50	3/31	4.6	279	0.51	49	1.73	58	1.87
Water <sup>b</sup>	9	3	3/31	4.8	175	1.62	33	1.58	44	1.33
ATP <sup>a</sup>	10	18	4/1	4.0	137	1.26	27	1.11	33	1.40
Water <sup>b</sup>	11	3	4/1	4.3	123	1.11	28	1.00	40	0.75
Water <sup>b</sup>	12	20	4/2	2.9	98	1.28	27	0.93	38 <sup>c</sup>	1.05
Water <sup>b</sup>	13	60	4/5	2.0	90	1.07	27	1.00	38	1.00
35% HNO <sub>3</sub> <sup>d</sup>	14	4	4/7	1.4	55	1.64	22	1.27	33	1.15
Water <sup>b,d</sup>	15	5	4/8	1.6	55	1.00	11	2.00	22	1.50
Water <sup>b</sup>	16	16	4/9	1.2	52	1.06	4	2.75	8	2.75
Water	17	3	4/9	2.2	46	1.13	8	0.50	8	1.00
Water <sup>b</sup>	18	60	4/12	1.8	36	1.28	7	1.14	7	1.14
Water <sup>b</sup>	19	2	4/12	0.9	36	1.00	8	0.88	6	1.17
5% HNO <sub>3</sub> <sup>d</sup>	20	22	4/14	0.2	29	1.24	6	1.75	5	1.20
Overall						23.8		22.0		25.0

<sup>a</sup>10% sodium hydroxide—1.5% sodium tartrate—1.5% hydrogen peroxide.

<sup>b</sup>Heated to about 100°C for 1 hr.

<sup>c</sup>Water emptied from heat exchanger outside of tube sheet.

<sup>d</sup>Entire system, about 500 liters, filled with solution; 200 liters used in all other cases.

acid treatment, the decontamination factor in the fuel solution cell was high; with the first 5% nitric acid treatment there was a large decontamination factor in the other two cells although the activity in the fuel solution cell was increased; and large decontamination factors were obtained when a reagent was heated for the first time and when the reactor system was completely filled for the first time.

#### 4.1 Individual Reagent Decontamination

Preliminary Water Wash. None of the earlier water washes were sampled or analyzed, and it is not known what fission products they contained. The last preliminary water wash, before the actual decontamination was started, contained fairly large amounts of cerium, iodine, and strontium, and some barium, lanthanum, and other rare earths. Large amounts of strontium and cerium remained in the system.

First 35% Nitric Acid. With the first 35% nitric acid (No. 1) wash, decontamination factors were 2.25 for the fuel solution cell and about 1 for the other two cells. This indicates that either the other two cells were not decontaminated by nitric acid or sufficient contamination was transferred into them from the fuel solution cell to maintain their activity nearly constant. This reagent removed approximately 1000 curies each of barium, lanthanum, and cerium, together with significant amounts of strontium and zirconium. This represents almost complete removal of barium and lanthanum. Except for zirconium, these are the fission products most likely to precipitate in the HRE system.

The 60-r/hr reading of the 25-ml sample taken just before the nitric acid was dumped (Table 1) is more than 10 times that of any other reagent, probably because of the high gamma activity of the  $Ba^{140}$ - $La^{140}$  pair and the high energy of the lanthanum gamma radiation.

The nitric acid may have dissolved some of the corrosion scale, since it was pink when removed from the reactor.

Water Wash Following First 35% Nitric Acid. The water wash (No. 2) that followed the first 35% nitric acid removed significant amounts of zirconium, niobium, and ruthenium. Apparently dilute acid, formed by the water wash plus the concentrated nitric acid heel, was more effective than concentrated acid for these fission products. The water washes (Nos. 9 and 15) following other acid treatments were also more effective for niobium and zirconium than the acid solution was.

The solution contained a small amount of suspended solids when removed from the system.

First 5% Nitric Acid. The first 5% nitric acid (No. 8) treatment produced marked changes in all cell radiation levels, nearly doubling the level in the fuel solution cell and halving it in the reactor and heat exchanger cells. The high reading in the fuel solution cell may have resulted from a transfer of activity into the cell, possibly from solids deposited on the filter, or merely from some of the solution being left in the cell near the probe when the system was drained.

Large amounts of cerium, zirconium, strontium, and trivalent rare earths were removed by this solution. The presence of such easily removed fission products as trivalent rare earths at this stage of decontamination suggests that a new source of fission products was contacted, for example, a plugged tube.

The used solution contained large amounts of solids that had the appearance of ordinary iron rust; this material was also found in all subsequent reagents in smaller amounts. It is possible that the dilute nitric acid reacted with and loosened part of the corrosion film, which carried large amounts of radioactivity; and it was the transfer of these solids, first to the fuel solution cell and finally out of the system, that caused the

changes in cell readings. The presence of this material, also, indicated that a plugged tube somewhere in the system may have opened. It is apparent that the decontaminating reagents did attack the corrosion film, but after the decontamination was completed, there was still a fairly heavy oxide coating on the inner surface of the system.

First Heated Solution. The water wash (No. 8) following the first 5% nitric acid treatment was the first reagent that was heated. Decontamination factors were 1.38 in the reactor cell, 1.62 in the fuel solution cell, and 1.33 in the heat exchanger cell.

First Reagent That Filled the Fuel Solution Cell Equipment. The first reagent that filled the equipment in the fuel solution cell was the second 35% nitric acid solution (No. 14). With this reagent a decontamination factor of 1.64, larger than any after the first nitric acid treatment, was observed in the fuel solution cell. Since some of the apparatus in this cell had not previously been contacted with decontaminating reagents, the increased decontamination was to be expected.

Alkaline-Tartrate-Peroxide. Decontamination factors only slightly greater than 1 were obtained with alkaline-tartrate-peroxide (Nos. 3 and 10). However, the larger decontamination factors observed with the other reagents were obtained only when some precipitate or contaminant that could easily be removed was dissolved. Although less effective than nitric acid for easily removed contaminants, it was more effective for niobium, and the second treatment removed about as much as the first. It thus contributed significantly to the overall decontamination even though  $Zr^{95}$ - $Nb^{95}$  was originally only a small fraction of the total contamination.

Last 5% Nitric Acid. The last 5% nitric acid (No. 20) did not remove much activity, but most of the fission products had already been removed from the system by the time of this treatment.

#### 4.2 Overall Decontamination

The overall decontamination factor, including decay, was 20-22. The decontamination factor due to the reagents alone was probably about 15. Since cell activity levels were low enough after the last 5% nitric acid wash for dismantling to begin, the reactor was washed with water to remove the acid and dismantling was started. Further decontamination could have been achieved by continued treatment, but probably only with difficulty and with removal of more of the corrosion film.

The gross beta and gamma activities of the fuel decayed by a factor of about 1.5 during the month of decontamination. The limiting activity after decontamination was  $Zr^{95}-Nb^{95}$ , so that the half-life of the remaining contamination was between one and two months.

It is possible that decontamination of the portions of the cell that were treated with the reagents was greater than indicated by the decontamination factors obtained. Radiation from the dump tanks at the bottom of the fuel cell undoubtedly resulted in higher radiation readings than would have been obtained from the decontaminated equipment alone. Also, the bottom part of the fuel solution cell and all the equipment in it had apparently been contaminated on the outside and was therefore not properly decontaminated by the reagents. The contribution of this radiation to the cell activity readings was only a small fraction of the total at the beginning of the decontamination, but it may have been a significant proportion at the end, after the cell activity levels had been reduced.

#### 5.0 RADIOACTIVITY REMAINING IN THE HRE AFTER DECONTAMINATION

Specimens of types 347 and 304L stainless steel, zirconium, and of titanium and type 347 specimen holders were in the reactor during operation and decontamination.\* When removed, the stainless steels

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\*Extensive studies on the nature and extent of corrosion were made by the Corrosion Group of the Reactor Experimental Engineering Division, ORNL.

were covered with a fairly heavy, loosely adhering rust-colored deposit, with no evidence of the blue-black oxide coating that forms on stainless steel under HRE operating conditions. The holders had an activity of about 25 r/hr/cm<sup>2</sup> at nearly contact by cutie-pie measurement. An alternate treatment with 35% nitric acid and alkaline-tartrate-peroxide gave a decontamination factor of only 1.1 per cycle. The zirconium and titanium were covered with a uniform oxide film, blue colored on zirconium, which showed no evidence of attack by the decontaminating reagents. Electrolytic descaling decontaminated the type 304L stainless steel by a factor of 80 but the other metals only slightly. Descaling with 1% oxalic acid decontaminated the type 347 stainless steel holders by a factor of 100 without serious corrosion. Analysis of the electrolytic descaling solution showed that more than 99% of the activity remaining in the scale on the metals except zirconium after the decontamination of the HRE system was due to Zr<sup>95</sup>-Nb<sup>95</sup>. The zirconium scale contained some cerium.

The corrosion specimens were located at each end of an elbow between the core and the heat exchanger in the fuel circulating system. The specimens were rectangular sheets mounted parallel to the fuel flow, across the diameter of type 347 stainless steel sleeves that slipped into the ends of the elbow. One set was in the system throughout operation and decontamination, and the other only during operation with the last fuel solution, which was for most of the power operation, and decontamination. Since they were in the heat exchanger cell rather than the reactor cell, they were exposed only to the delayed neutron flux of the fuel. The scale on the specimen holders was heavier and the radioactivity per unit area was about five times that of the type 347 specimens, but the relative amounts of fission products were the same on all stainless steel samples. The higher specific activity of the holders may have been due to a lower flow velocity around the edges of the pipe than in its interior.

## 5.1 Descaling of Corrosion Specimens and Holders

Electrolytic Descaling of Specimens.\* When electrolytically descaled, the type 304L stainless steel specimens were decontaminated by a factor of about 80, but the type 347 specimens by a factor of only 2 (Table 2). Titanium was decontaminated by a factor of only 1.5 to 2, and zirconium by a factor of only about 1.1. The blue oxide coating on zirconium was not visibly attacked by the descaling treatment. If these metals are used in a homogeneous reactor system, they may offer a serious decontamination problem unless some method is devised to descale them chemically without corroding the rest of the system.

The electrolyte for the descaling was 5% sulfuric acid inhibited with Rhodine-77. After current had been passed for a few minutes, the samples were brushed under water to remove any loosely adhering scale. The radioactivity level of each specimen was measured with a cutie-pie before and after the descaling, and the amounts of fission products removed by descaling were determined by standard radiochemical analyses.

Chemical Descaling of Holders. A decontamination factor of the order of 100 was obtained by chemical descaling; the time and temperature required for this amount of decontamination varied with the reagent. With 1% oxalic acid at 80°C, decontamination factors of the order of 100 were obtained in about 30 hr (Fig. 2). The rate of decontamination was roughly proportional to the oxalic acid concentration up to about 5%, the highest concentration tested. The same decontamination was obtained with 1.0 M hydrochloric acid—1.4 M sulfuric acid—0.2 M hydrogen peroxide inhibited with alkyl pyridines HB in 3 to 5 hr at 80°C; in a few minutes with concentrated sulfuric acid at 150°C; and in 6 days with 3-20 reagent (3% hydrofluoric acid in 20% nitric acid) at room temperature.

A piece of a specimen holder (about 120 cm<sup>2</sup> surface area) that had been treated for a week with 1% oxalic acid at about 70°C showed some corrosion, but microscopic examination showed it to be a more

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\*Work of A. R. Olsen, Reactor Experimental Engineering Division.

Table 2. Decontamination of HRE Corrosion Specimens  
by Electrolytic Descaling<sup>a</sup>

Specimen	Radiation Level (mr/hr) <sup>b</sup>	
	Before Descaling	After Descaling
Set 1 <sup>c</sup>		
347 stainless steel	1700	800
	4600	700
Ti	1600	680
	1500	680
	1850	700
	1850	850
	1400	780
Set 2 <sup>c</sup>		
Zr	3400	3100
	3700	3100
304L stainless steel	900	10-11
	1050	14-30
Ti	800	550
	1000	690
	1150	710

<sup>a</sup>Descaled electrolytically in 5% sulfuric acid inhibited with Rhodine-77. Readings given are average for the two sides of specimen.

<sup>b</sup>Cutie-pie reading at about 0.5 in. through several thicknesses of aluminum foil which absorbed beta radiation.

<sup>c</sup>Set 1 remained in reactor throughout operation and decontamination; set 2 was in the system only during operation with the last fuel solution, which was for most of the power operation, and decontamination.

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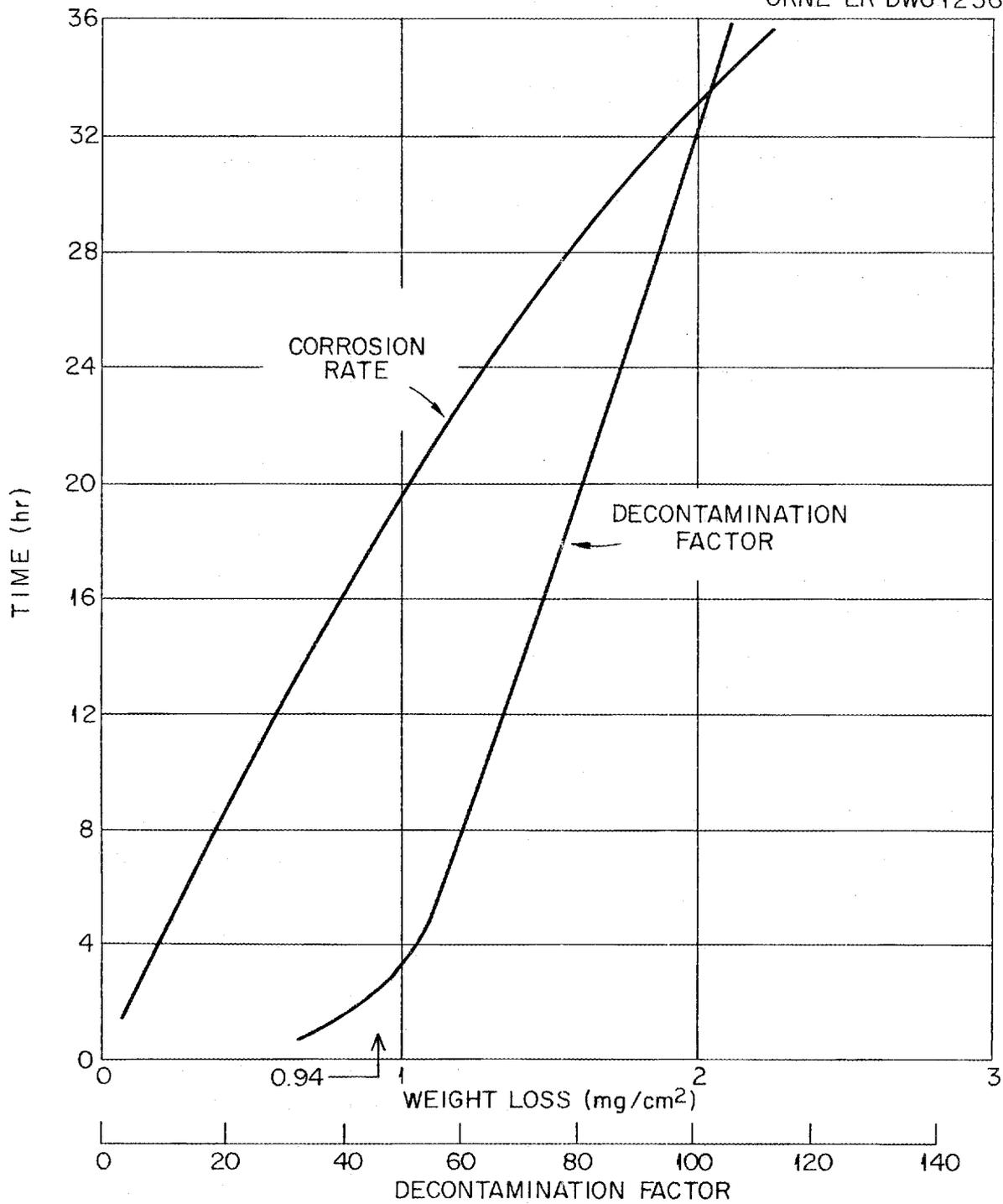


Fig. 2. Corrosion Rate and Decontamination Factor of a Portion of Type 347 Stainless Steel Corrosion Specimen Holder from HRE, Decontaminated with 1% Oxalic Acid at 80°C. Total surface area = 1 cm<sup>2</sup>; corrosion rate = 0.033 mg/hr (0.0017 mil/hr).

general attack than the intergranular corrosion obtained with strong mineral acids. In this experiment decontamination factors were 10 in 1 day, 25 in 2 days, 60 in 3 days, and 160 in 7 days. For large-scale use, oxalic acid would probably be preferred to mineral acids, although requiring more time for the same decontamination, because of being less corrosive. In addition, the oxalic acid could be destroyed by heat, and the fission products concentrated into a small volume for lower cost storage.

## 5.2 Descaling of Unmodified Film

The oxide film in the HRE had been modified considerably by the decontamination treatment. In order to investigate the ability of oxalic acid to descale the unmodified corrosion film, parts of an in-pile corrosion loop were contacted with 5% oxalic acid at 80°C for 30 hr. The scale was removed, although more slowly than from parts of the HRE system. The parts of the loop were also decontaminated from fission products, but there was a considerable amount of gamma activity remaining, resulting from neutron activation of chromium, iron, and cobalt. Initially, fission products contributed about 90% of the gamma activity, and after the descaling they contributed only about 25%, the other 75% being from chromium, iron, and cobalt. The most important fission product contaminants were niobium and zirconium, but there were also some ruthenium and a very little cerium. The decontamination and corrosion measurements were not completed, but the decontamination factor for fission products appeared to be near 40 and the overall corrosion between 0.1 and 0.2 mil.

The action of oxalic acid appeared to be quite different from that of mineral acids. The mineral acids presumably do not dissolve the scale but rather remove a layer under it, permitting it to fall off and leave a bright metal surface. With oxalic acid, however, the red oxide is slowly replaced by a gray coating which remains on the surface. Some solids fall off the surface and are not dissolved.

This gray oxide was identified as  $\gamma\text{-Fe}_2\text{O}_3$  by electron diffraction examination.\* The same diffraction pattern was observed in new type 347 stainless steel exposed to hot oxalic acid and in a piece of the corrosion specimen holder that was descaled with oxalic acid. Another pattern observed with the latter sample is not a known pattern of the constituents of stainless steel or of uranium and was not identified.

### 5.3 Contamination on Specimens and Holders Removed from the HRE

Results of analyses of the electrolytic descaling solutions showed that the radioactivity in the scale on each metal except zirconium after the mild decontamination was practically all due to  $\text{Zr}^{95}\text{-Nb}^{95}$  (Table 3). There was some cerium contamination on the zirconium. No evidence was found for neutron activation of the metals. The Nb/Zr activity ratio on the stainless steel was about 5/1, a value considerably higher than the equilibrium value obtained by zirconium decay. On titanium and zirconium metals, however, the Nb/Zr ratio was about 1.5, approximately the theoretical value for the HRE fuel at the time of analysis. It therefore appears that the corrosion film on zirconium and titanium is relatively impervious to attack by decontaminating reagents. On the other hand, the corrosion film on the stainless steels is more open to attack by the decontaminating reagents, resulting in selective removal of zirconium. On the basis of the activity on the type 347 stainless steel specimens, about 1% of the calculated amount of niobium produced by the HRE and 0.3% of the zirconium was left in the system (about  $10^5 \text{ cm}^2$  surface area), and, on the basis of the activity on the specimen holders, about five times as much was left in the system.

Table 3. Fission Products Removed by Electrolytic Descaling  
of Corrosion Specimens<sup>a</sup>

Specimen	Activity x 10 <sup>-7</sup>			Percentage of Total			
	Gross β (c/m)	Gross γ (c/m)	Total (d/m)	Zr	Nb	Ce	TriRE
Zr	5.8	49.7	204	40.1	55.3	4.4	0.2
Ti	18.2	185	774	38.0	61.7	0.1	0.2
304L stainless steel	17.9	247	987	16.6	83.1	0.2	0.1
347 stainless steel	36.5	523	2050	17.2	82.3	0.4	0.1

<sup>a</sup>Average amount of fission product removed from each sample of the particular metal (12.7 cm<sup>2</sup> area). Ruthenium was not determined because of an accident in sample preparation. The ruthenium contribution was probably not more than 1 or 2% of the total activity. Analyzed 5/20/54.

## 6.0 DISTRIBUTION OF THE HRE-PRODUCED FISSION PRODUCTS

Of the fission products found by analysis of the HRE fuel and decontaminating reagents, most of the cesium and of the trivalent rare earths except cerium and lanthanum were in the fuel; about half the cerium and strontium and most of the zirconium, niobium, ruthenium, iodine, barium, molybdenum, and lanthanum were in the decontaminating solutions. The gross beta activity of all spent decontaminating solutions was less than that of the fuel at the same time, but the gross gamma activity of several was higher, which indicates the tendency of gamma-emitting fission products to precipitate from the circulating fuel solution. Cesium was the only strong gamma-emitting fission product found in the fuel. Only a small fraction of the plutonium formed was found in the fuel. Insufficient data were obtained to determine the distribution of neptunium and of tellurium and other short-lived fission products.

Since the fission products in the fuel removed from the reactor at the end of operation were determined at various times and long-time decay corrections are difficult to make, material balances were calculated from analyses of the spent decontaminating reagents and of two fuel samples taken from the reactor during operation (see appendix for details). The sums of the amounts of fission products found in these solutions were compared with the amounts calculated to have been produced during operation. Material balances were within  $\pm 20\%$ , which is about the best that can be expected under such conditions, for cesium, cerium, and the trivalent rare earths except cerium (Table 4). Ruthenium, niobium, and molybdenum balances were low. Balances for the other fission product determined were around 50%. The low balances may have been caused by the following facts:

1. The two samples may not have been at equilibrium when removed from the reactor. They were analyzed in different laboratories, and some of the discrepancies may have resulted from

Table 4. Distribution of the HRE-Produced  
Fission Products

Fission Product	% of Calculated Total Activity Found				
	In Decontaminating Reagents <sup>a</sup>	In HRE Fuel Sample 1	In HRE Fuel Sample 2		
			Whole Sample	Solids	Supernatant
TriRE except Ce <sup>b</sup>	14	100	70	0.2	70-100
Ce	64	81	44	0.2-2	42
La <sup>c</sup>	43	---	4	< 1	~ 3
Zr	45	~1	< 1	0.2	0.3
Nb	20	> 1	~ 1	0.4	0.4
Ru	11	0.5	0.4	0.2	0.2
Ba <sup>d</sup>	54	~ 10	~ 2	4	10
Sr	34	35	14	13	< 1
Mo	---	30	9	---	---
Cs	0.1	---	120	0.5	120
I	45	11	12	0.5	10

<sup>a</sup>Summation of the percentages of the calculated amounts of fission products in all decontaminating reagents.

<sup>b</sup>Including lanthanum.

<sup>c</sup>Lanthanum was calculated from the trivalent rare earth gamma count.

<sup>d</sup>Barium analytical values in the fuel varied greatly, appearing to increase with time.

differences in analytical techniques.

2. The volumes of the spent decontaminating reagents were not measured accurately and no account was taken of the heel left in the system each time a solution was dumped.

3. Small amounts of solids, similar to the corrosion scale in the decontaminating reagents, were suspended in the fuel and they carried about as much ruthenium, barium, niobium, and zirconium as the fuel solution did. These solids were difficult to dissolve, and the leaching procedure finally adopted may not have removed all the fission products from the solids before the solutions were analyzed.

4. Volatile fission products may have left the solution during operation. A mist trap in the HRE gas system caught large amounts of volatile fission products, notably the rubidium and strontium daughters of krypton. Other isotopes may have been lost in the same way, including daughters of xenon and possibly ruthenium and iodine.

5. The samples may not have been representative.

6. Some zirconium and niobium remained in the reactor after the decontamination.

Barium appeared to increase with time in the fuel samples, indicating that barium sulfate may have dissolved slowly upon standing. Lanthanum was always found with barium, probably as a result of barium sulfate precipitating in the system and partially decaying to lanthanum, which could not escape from the crystal until the barium sulfate was dissolved.

The low percentage of cerium in the decontaminating reagents compared to the other rare earths would not have been expected. Although it is the least soluble rare earth in the trivalent form, its solubility probably was not reached in the HRE. However, the relative ease of its oxidation to the tetravalent state and subsequent hydrolysis may have affected its distribution.

The behavior of molybdenum and its daughter, technetium, was of some interest because of the possible high neutron cross-section of technetium. Molybdenum had decayed to a small fraction of the gross activity before the decontaminating reagents could be analyzed, and only about 30% of the calculated amount of molybdenum was found in the fuel. The calculations for such a short-lived isotope as molybdenum are subject to large error, and analysis for technetium in such solutions is difficult because of its low specific activity compared to the gross activity of the HRE solutions. However, indications were that the amount of technetium in the fuel was at least 10 times that in the decontaminating reagents.

A large proportion of the plutonium produced by the HRE was found in the decontaminating reagents. The exact amount of plutonium produced is difficult to calculate because of the neutron energy spectrum of the reactor, but it was calculated to be 74 mg (11 mg by thermal capture and 63 mg by resonance capture) on the basis of 660 Mw-hr of operation.\* In all solutions the amounts were measured as only a few counts per minute of activity, and determinations are therefore subject to considerable error. About 20% of this theoretical amount of plutonium was found in the fuel (11.8 mg in solution and 3.4 mg in fuel solids). Seventy-six milligrams of plutonium was found in the decontaminating reagents.

Gaseous precursors, if of sufficiently long half-life, could decrease the fission products in the system. The water from the gas recombiner could be the source of a fairly pure fraction of fission products. Daughters of gaseous precursors with half-lives long enough to be carried from the fuel system in the gas stream---but short enough to decay before reaching the recombiner-condenser---may dissolve in the condensate.

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\*Personal communication from P. M. Wood, Reactor Experimental Engineering Division, ORNL.

## 7.0 APPLICATION OF RESULTS TO LARGER HOMOGENEOUS REACTORS

Many fission products in a homogeneous reactor will reach a solubility limit and precipitate in the fuel, for example, the rare earths, barium, and strontium. Results of the decontamination studies on the HRE indicate that other fission products— notably zirconium, niobium, ruthenium, and iodine—will probably leave solution, either precipitating or adsorbing on the walls of the equipment. This latter group may be tied up with the corrosion film, which, apparently, continuously flaked off stainless steel in the HRE, where operation was intermittent. All these fission products will probably be found in large quantities in the solids removed from the HRE fuel. Cesium is the only one considered here that will almost certainly stay in solution in the fuel. The others will build up to some quantity less than that expected from the fission yield, the amount being determined by solubility, adsorption, volatilization, or any process by which fission products can be removed from the fuel.

The decontamination achieved in the HRE may not be sufficient for decontamination of a large reactor system for maintenance or modification. It is doubtful that appreciably more decontamination could be achieved without descaling the system. Therefore the whole problem of descaling and corrosion resulting from descaling should be investigated. By descaling the HRE, a decontamination factor of 2000 instead of 25 probably could have been obtained. Oxalic acid solutions show promise with respect to decontamination and ease of storage, but corrosion by oxalic acid has not been investigated in detail. Until more is known about the corrosion problem, decontamination would be limited to the same sort of procedure used for the HRE. This could probably be improved in efficiency by using more nitric acid treatments at first to dissolve any fission product precipitates that may be in the system, such as rare earths and barium, until nitric acid has little effect. Then alkaline tartrate-peroxide should be used alternately with nitric acid in the hope that it might remove some of the niobium and zirconium left in the system.

In the HRT there may be only small amounts of fission product precipitates in the reactor system since, unlike in the HRE, solids will be continuously removed from the fuel.

For a homogeneous power reactor the problem of decontamination would be more concerned with time. For example, shutdown of a 100-Mw power station, because of equipment failure, would cost about a quarter of a million dollars per day just for replacement of the power normally produced. Thus, the system must be repaired immediately, and a decontamination treatment would be used only if it could effect a large reduction in radioactivity in a few hours, at most. The only hope of this would be by descaling. A decontamination treatment such as that used for the HRE could not be considered.

#### 8.0 CONTAMINATION AND DECONTAMINATION OF TYPE 347 STAINLESS STEEL COUPONS

In a few preliminary scouting experiments to determine what might be expected in the HRE, the most important initial contaminant on type 347 stainless steel coupons exposed to HRE fuel at room temperature was found to be iodine;  $Zr^{95}$ - $Nb^{95}$ , ruthenium, and, in one experiment, cerium were also present in significant amounts. No evidence of any rare earth other than cerium was found. Gross gamma decontamination factors for the first treatment of the coupons were about 10 with alkaline-tartrate-peroxide for a day and 4 with nitric acid for a few hours; factors dropped to 1-2 in subsequent treatments with any reagent. Three cycles with 8 M nitric acid followed by alkaline-tartrate-peroxide gave a gross gamma decontamination factor of about 100.

For the experiments 1-cm<sup>2</sup> coupons of type 347 stainless steel were immersed at room temperature in a sample of HRE fuel solution withdrawn from the reactor while it was operating, in early February, 1954. After being in the solution for one to three days, the coupons were removed, rinsed with water, and then immersed for various lengths

of time in 5% nitric acid, 35% nitric acid, 5% oxalic acid, or 10% sodium hydroxide—1.5% sodium tartrate—1.5% hydrogen peroxide. The radiation levels were determined with a cutie pie or a scintillation spectrometer after each treatment, and the coupon was then immersed in one of the other reagents.

For removing iodine, nitric acid was the most effective reagent, and there was little difference between 5% and 35%. Water gave some decontamination. The alkaline-tartrate-peroxide reagent gave poor iodine decontamination after the first treatment.

For removal of  $Zr^{95}$ - $Nb^{95}$ , alternate treatment with 35% nitric acid and alkaline-tartrate-peroxide was most effective. Five percent nitric acid was less efficient than 35%. With all reagents the ruthenium decontamination factor was as large as or somewhat larger than that of  $Zr^{95}$ - $Nb^{95}$ , so that ruthenium removal was never a serious problem. Cerium was also removed with less difficulty than zirconium and niobium.

## 9.0 APPENDIX

### 9.1 Counter Efficiency Factors

Factors for converting counts per minute (cpm) given by standard counting procedures to disintegrations per minute (dpm) for a number of fission products were supplied by S. A. Reynolds of the Analytical Chemistry Division, Oak Ridge National Laboratory (Tables 5 and 6). The factor is the quantity by which the counting rate must be multiplied to obtain an approximate disintegration rate. For beta counts the counting rate referred to is that obtained by making a radiochemical separation and counting the fission product activity on the second shelf of the standard ORNL Geiger-Mueller beta counter. For gamma counts the counting rate referred to is that obtained with an analytical scintillation counter with a 5-g/cm<sup>2</sup> lead absorber. The factor includes geometry, self-scattering, and absorption corrections.

The factors for mixtures of fission products (such as rare earths, Ce<sup>141</sup>-Ce<sup>144</sup>-Pr<sup>144</sup>, or Ru<sup>103</sup>-Ru<sup>106</sup>-Rh<sup>106</sup>) depend upon bombardment and decay time. The factors for such mixtures were estimated from decay and absorption data. The errors in the factors are not known, but are probably not more than 10 to 15% for such isotopes as barium, iodine, strontium, and neptunium. These factors were used to calculate the values reported in the tables of this report from the results of radiochemical analyses.

### 9.2 Calculation of Amounts of Fission Products Produced by the HRE

An approximate calculation was made on the Oracle of the amounts of important fission products produced in the HRE that were present at various times after its shutdown. A similar calculation was made for the fission products in the two fuel samples removed from the reactor before it was shut down. The calculations were made as follows:

Several assumptions and approximations had to be made. Examination of the HRE log showed that it operated, for the most part, for several periods of a few days each and did not operate between these periods. It was assumed that each period could be treated as if all the power had been produced at the median date of the period. For short-lived isotopes (those with half-lives comparable to the length of the operating period) and for short decay times, when such isotopes are important, this assumption is certainly a source of error. Most of the decay times of interest were sufficiently long that such short-lived isotopes were not significant. However, if the problem should be done again, some of the longer operation periods, especially toward the end of the operation, should be broken up into smaller increments for greater accuracy.

There is some question as to just how much of the HRE operation should be included in the calculation, since it operated for about a tenth of its total power with a different fuel solution from that used in the later work. The first fuel became contaminated chemically and was removed and processed early in 1953. At the same time the reactor system was thoroughly washed, but it was not decontaminated to any great extent. Since this operation was over a year before the times of interest, the fission product contribution from this early period should be very small except for long-lived fission products, of which the most important (cesium) certainly was removed with the first fuel. For the purposes of this calculation, operation with only the last fuel solution was considered. The power values given in Table 7, which were obtained from the HRE records and may be low by 10 to 20%, were used in the calculations.

The calculations were made for the isotopes listed in Table 8, using the yield and decay constant given. In addition, the constants for several shorter lived fission products— $\text{Ce}^{143}$ ,  $\text{Eu}^{156}$ ,  $\text{Sm}^{153}$ ,  $\text{Sm}^{151}$ ,  $\text{I}^{149}$ ,  $\text{Sr}^{91}$ ,  $\text{Y}^{93}$ ,  $\text{Zr}^{97}$ , and  $\text{I}^{133}$ —were coded for the Oracle and the amounts of these isotopes were calculated. Calculation by this procedure of such short-lived fission product constants is, however, subject to large error, and none of these values are significant except those for fuel sample 1 on February 4.

Table 5. Counter Efficiency Factors for Elements with Only One Important Fission Product Isotope

Isotope	Beta Factor	Gamma Factor
Ba-140	13.5	16.7
Cs-137	---	5.2
I-131	17.5	7.6
La-140	---	2.7
Nb-95	185	4.5
Zr-95	26	4.7
Mo-99	13	24
Sr-89	12	---
Np-239	10	---

Table 6. Beta Counting Factors for Rare Earths, Cerium, Ruthenium, and Tellurium in HRE Fuel

Date	Ruthenium	Cerium	Trivalent Rare Earths except Cerium	Tellurium
2/4/54	38	14	12	10
2/12	38	14	12	10
3/1	31	13	13	13
3/30	24	12	13	13
4/15	22	11	13	---
4/30	20	11	13	---
5/15	19	10	13	---
5/31	17	10	13	---

Table 7. Summary of the ERE Operation

n	Period of Operation	Median Date	Power (Mw-hr)	Moles U <sup>235</sup> fissioned	Time from Median Date to Date of Interest (days)
					<u>To 2/4/54</u>
1	11/20-11/25/53	11/23/53	93.2	0.0182	73
2	12/1-12/9	12/4	79.3	0.0155	62
3	12/18-12/23	12/20	119.1	0.0232	46
4	1/4-1/8/54	1/6/54	38.1	0.0075	29
5	1/18-1/26	1/22	107.2	0.0209	13
6	1/28-1/29	1/29	22.0	0.0043	6
Fuel sample 1 removed for analysis					
7	2/4-2/6	2/5	11.2	0.0022	---
Fuel sample 2 removed for analysis					
					<u>To 3/24/54</u>
8	2/16-2/18	2/17	16.0	0.0031	35
9	2/23-2/25	2/24	16.0	0.0031	28
10	3/2-3/4	3/3	16.0	0.0031	21

Table 8. Yields and Decay Constants for Some Fission Products

Isotope	A	Z	Fission Yield	Half-life (days)	Decay Constant (days <sup>-1</sup> )
Mo	99	42	0.062	2.79	0.248
Ru	103	44	0.037	39.8	0.0174
Ru	106	44	0.0040	365	0.0019
Zr	95	40	0.0677	65	0.01066
Sr	89	38	0.046	53	0.0131
Sr	90	38	0.0610	7,300	0.000095
I	131	53	0.0297	8.14	0.0851
Ba	140	56	0.0607	12.8	0.0541
Ce	141	58	0.046	33.1	0.0209
Ce	144	58	0.0576	282	0.00246
Cs	136	55	0.00006	13	0.0535
Cs	137	55	0.0619	13,500	0.0000513
Y	91	39	0.0624	57	0.01216
Pr	143	59	0.061	13.7	0.0506
Nd	147	60	0.026	11.3	0.0613
Pm	147	61	0.026	950	0.000729
Pm	148	61	0.0164	48	0.0144

Since niobium did not fit the specific Oracle code use it was calculated by hand from the data of Table 10. Lanthanum<sup>140</sup> was assumed to be in equilibrium with Ba<sup>140</sup>. Since the decay constant of lanthanum is much greater than that of barium, the activity of lanthanum is approximately 1.15 times that of barium.

### 9.3 Detailed Tables of Data

Tables 9 through 14 are tables of detailed data.

Table 9. Fission Products Found in Decontamination Reagents

Reagent	Reagent No.	Fission Product Activity ( $10^7$ d/m/ml) <sup>a</sup>								
		TriRE <sup>b</sup> except Ce	Ce	La	Ba	Sr	Zr	Nb	Ru	I
Date analyzed:		3/24								
Water	1	150	310	60	60	114	0.3	0.4	0.1	120
35% HNO <sub>3</sub>		975	1,250	950	1,000	250	260	20	16	40
Date analyzed:		3/29								
Water	2	290	450	90	110	200	410	110	93	5.0
ATP <sup>c</sup>	3	27	20	1.3	2.0	6.0	141	220	28	60
Water	4	13	12	1.0	1.5	4.5	23	48	12	3.0
Date analyzed:		4/30	4/20	4/13	4/13	4/20	4/20	4/30	4/30	4/13
Water	5	4.6	18	0.6	0.6	9.0	21	38	8.0	0.5
Water	6	3.1	14	0.4	0.3	5.0	25	21	3.0	0.4
Water	7	1.4	7.0	0.2	0.1	2.0	11	10	0.6	0.4
5% HNO <sub>3</sub>	8	155	600	20	18	348	400	53	50	0.1
Water <sup>d</sup>	9	155	800	18	16	322	375	200	8.0	0.4
ATP <sup>c</sup>	10	7.4	18	1.0	0.5	6.0	200	195	7.0	0.3
Water <sup>d</sup>	11	1.9	10	0.2	0.1	2.0	85	63	14	0.4
Water <sup>d</sup>	12	1.1	5.0	0.1	0.1	1.3	63	75	9.0	0.3
Water <sup>d</sup>	13	1.0	5.0	0.1	0.1	1.1	49	44	5.0	0.3
35% HNO <sub>3</sub> <sup>e</sup>	14	3.1	15	0.4	0.4	4.0	90	38	9.0	0.1
Water <sup>d, e</sup>	15	3.5	14	0.4	0.4	5.0	100	41	7.0	0.2
Water <sup>d</sup>	16	3.4	13	0.4	0.4	4.0	95	38	7.0	0.2
Date analyzed:		5/6								
Water	17	---	1.0	0.03	0.04	0.6	3.0	3.1	0.8	---
Water <sup>d</sup>	18	---	2.1	0.1	0.1	1.4	5.2	2.4	0.7	---
Water <sup>d</sup>	19	---	1.5	0.06	0.1	1.0	2.3	1.7	0.3	---
5% HNO <sub>3</sub> <sup>e</sup>	20	---	3.6	0.1	0.1	2.7	8.4	2.8	0.7	---
Water		---	0.9	0.02	0.04	0.7	1.2	1.0	0.3	---

<sup>a</sup>The numbers were calculated from the results of standard radiochemical analyses, which are in units of counts per minute per milliliter, by means of counter efficiency factors tabulated in the appendix. The numbers in the table in  $10^7$  d/m/ml are approximately equal to the number of curies of the isotope in the entire solution, differing by a factor of 0.91. The total activity of the solution for the 200-liter volumes would be the number in the table  $\times 2 \times 10^5 \times 10^7$ , or  $2 \times 10^{12}$  d/m/ml; a curie is  $2.2 \times 10^{12}$  d/m/ml.

<sup>b</sup>Lanthanum was calculated from the trivalent rare earth (TriRE) gamma count, assuming that all gamma activity was due to lanthanum. The trivalent rare earth value was calculated from the beta count, and represents all the rare earths except cerium.

<sup>c</sup>10% sodium hydroxide--1.5% sodium tartrate--1.5% hydrogen peroxide.

<sup>d</sup>Heated to about 100°C for 1 hr.

<sup>e</sup>Entire system filled with solution, to volume of about 500 liters. Otherwise, volume was 200 liters.

Table 10. Fission Products Found in HRE Fuel<sup>a</sup> Samples Removed during Operation

Values given for date of removal and various decay times

Isotope	Sample 1 <sup>a</sup>				Sample 2 <sup>a</sup>			
	2/4	2/12	3/1	3/30	2/16	2/24	3/25	5/7
Activity ( $10^7$ c/m/ml)								
Gross $\beta$	---	---	---	---	1,760	---	793	444
Gross $\gamma$	---	---	---	---	---	193	78.5	49
Activity ( $10^7$ d/m/ml)								
TriRE <sup>b</sup> except Ce	34,000	26,000	18,000	10,000	14,000	---	7,000	3,000
Ce	10,000	7,000	6,000	3,600	4,000	---	2,300	1,400
La <sup>b</sup>	---	---	---	---	---	220	26	10
Ba	780	710	410	110	82	71	102	1
Sr	2,400	2,200	1,800	1,100	840	---	547	270
Cs	---	---	---	---	---	80	71	75
Zr	240	40	50	50	30	34	75 <sup>c</sup>	16
Nb	220	100	40	30	---	37	45	84
Ru	30	30	20	10	18	---	11	8
I	700	320	60	---	---	234	9 <sup>c</sup>	---
Mo	1,700	230	3	---	64	68	0.3	---
Te	80	20	3	---	11	---	---	---

<sup>a</sup>See Sec. 9.2 for radiation history.

<sup>b</sup>Trivalent rare earths (including lanthanum, excluding cerium) calculated from beta count; lanthanum calculated from gamma count.

<sup>c</sup>Range of results calculated from beta and gamma counts = 44-107 for zirconium, 7-11 for iodine.

Table 11. Fission Products Found in Solids and in Supernatant of HRE Fuel Sample 2

Isotope	Analyzed 3/25		Analyzed 5/7	
	Supernatant	Solids	Supernatant	Solids
Activity ( $10^7$ c/m/ml)				
Gross $\beta$	760	12	460	10
Gross $\gamma$	52	23	31	20
Activity ( $10^7$ d/m/ml)				
TriRE <sup>a</sup> except Ce	6,000	14	3,380	11
Ce	2,100	13	1,428	71
La <sup>a</sup>	28	0.8	9	1
Ba	45	197	3	7
Sr		10	278	19
Cs	69	0.2	75	0.5
Zr	15 <sup>b</sup>	10	12	11
Nb	11	19	31	25
Ru	5	5	1	4
I	10	0.5	---	---
Mo	0.4	6 <sup>c</sup>	---	---

<sup>a</sup>Trivalent rare earths calculated from beta count (includes lanthanum); lanthanum calculated from TriRE gamma count.

<sup>b</sup>Range of results = 10-20; beta and gamma counts different.

<sup>c</sup>This value is unreasonably high and is probably in error.

Table 12. Calculated Amounts of Fission Products in HRE Fuel Samples Removed during Operation

Values given for date of removal and various decay times

Isotope	Sample 1				Sample 2			
	2/4 <sup>a</sup>	2/12	3/1	3/30	2/16	2/24	3/25	5/7
	Activity (10 <sup>7</sup> d/m/ml)							
Y-91	9,000	8,000	6,000	5,000	8,000	7,000	5,000	3,000
Pr-143	12,000	8,000	3,000	790	7,000	5,000	1,100	130
Nd-147	5,000	3,000	1,100	190	2,800	1,700	300	21
Pm-147	340	340	340	330	350	350	300	330
Pm-148	2,500	2,200	1,700	1,000	2,000	2,000	1,300	700
La-140 <sup>b</sup>	14,000	9,000	4,000	750	8,000	5,000	1,100	110
Total	43,000 <sup>a</sup>	31,000	16,000	8,000	28,000	21,000	9,000	4,000
Ce-141	8,000	7,000	5,000	2,600	7,000	6,000	3,000	1,300
Ce-144	2,400	2,300	2,200	2,100	2,400	2,300	2,000	2,000
Total	10,000 <sup>a</sup>	9,000	7,000	5,000	9,000	8,000	5,000	3,000
Ba-140	12,000	8,000	3,000	650	7,000	5,000	970	95
Sr-89	7,000	6,000	5,000	3,000	6,000	5,000	4,000	2,000
Sr-90	110	100	100	100	110	100	100	110
Total	7,000	6,000	5,000	3,000	6,000	5,000	4,000	2,000
Cs-136	4	2	1	0.2	2	2	0.3	0
Cs-137	59	59	59	59	60	60	60	60
Total	63	61	60	59	62	62	60	60
Zr-95	9,000 <sup>a</sup>	8,000	7,000	5,000	8,000	7,000	5,000	3,400
Nb-95	5,000	6,000	6,000	6,000	6,000	6,000	6,000	5,000
Ru-103	6,000	5,000	4,000	2,400	5,000	5,000	2,700	1,300
Ru-106	130	130	130	120	130	130	120	110
Total	6,000	5,000	4,000	2,500	5,000	5,000	2,800	1,400
I-131	6,000 <sup>a</sup>	3,000	660	56	2,000	1,200	110	3
Mo-99	6,000	770	11	0	750	100	0	0

<sup>a</sup>Date of removal. Certain short-lived isotopes—Zr-97, Sr-91, I-133, Ce-143, and several rare earths—contributed to the activity for such short decay.

<sup>b</sup>Lanthanum assumed in equilibrium with barium. Values for short decay should be lower.

Table 13. Calculated Amounts of Fission Products Produced by the HRE during Total Operation

Values given for various dates after shutdown

Isotope	Activity ( $10^7$ d/m/ml)						
	3/24	3/29	4/13	4/20	4/30	5/6	7/1
Ce-141	4,000	3,800	2,800	2,400	1,900	1,700	530
Ce-144	2,400	2,400	2,300	2,300	2,200	2,200	1,900
Total	6,000	6,000	5,000	5,000	4,000	4,000	2,400
Y-91	6,000	6,000	5,000	4,000	4,000	4,000	1,800
Pr-143	2,700	2,100	990	700	420	310	18
Nd-147	900	670	270	170	94	70	2
Pm-147	400	370	370	370	370	360	350
Pm-148	1,600	1,500	1,200	1,100	950	870	390
La-140 <sup>a</sup>	2,900	2,200	980	670	390	280	14
Total	15,000	13,000	9,000	7,000	6,000	6,000	2,600
Zr-95	6,000	6,000	5,000	5,000	4,000	4,000	2,300
Nb-95	7,000	7,000	6,000	6,000	6,000	6,000	4,000
Total	13,000	13,000	11,000	11,000	10,000	10,000	6,000
Ru-103	4,000	3,000	2,500	2,200	1,900	1,700	640
Ru-106	140	140	130	130	130	130	110
Total	4,000	3,000	2,600	2,300	2,000	1,800	800
Ba-140 <sup>a</sup>	2,500	1,900	850	580	340	240	10
Sr-89	5,000	4,000	3,000	3,000	2,800	2,600	1,200
Sr-90	120	120	120	120	120	120	120
Total	5,000	4,000	3,000	3,000	2,900	2,700	1,300
Cs-136	0.8	0.6	0.3	0.2	0.1	0.1	---
Cs-137	66	66	66	66	66	66	66
Total	67	67	66	66	66	66	66
I-131	630	410	110	60	30	20	<1
Mo-99	66	19	0.4	<0.1	---	---	---

<sup>a</sup>Lanthanum reaches equilibrium with barium. Values for lanthanum are 1.15 times those for barium.

TABLE 14. Cell Activity Levels

Date	Time	Activity (r/hr)			Remarks
		Fuel Cell	Reactor Cell	Heat Exchanger Cell	
2-7-54	10:10 AM	24,100	6,500	11,700	Fuel in outer dump tanks and system rinsed; reactor had operated at 400 kw for ~ 3 days
2-8-54	9:00 AM	21,900	5,630	9,900	
2-9-54	10:30 AM	18,300	7,050	10,100	
2-10-54	12:30 PM	17,600	4,320	7,390	
2-11-54	10:00 AM	17,300	4,150	6,900	
2-12-54	8:00 AM	17,100	3,940	6,450	
2-15-54	7:30 AM	15,100	3,390	5,470	
2-16-54	10:00 AM	34,200	32,500	24,900	Reactor had operated at ~ 400 kw for ~ 8 hr
2-17-54	9:30 AM	39,600	33,100	26,200	Reactor had operated at ~ 400 kw for ~ 31 hr
3-3-54	7:30 AM	47,000	33,900	27,300	Reactor had operated at ~ 400 kw for 40 hr
3-3-54	8:40 AM	17,000	3,170	2,790	Diluted core at 7:50 AM
3-3-54	9:50 AM	10,400	1,915	1,890	Dilute fuel still circulating
3-3-54	12:05 PM	5,300	1,050	1,285	Fuel drained to outer dump tanks at 10:00 AM
3-3-54	3:07 PM	3,720	945	902	Inner dump tanks were rinsed at 1:00 PM and drained to outer dump tanks; fuel heat exchanger full
3-4-54	8:00 AM	1,970	666	629	
3-4-54	12:35 PM	1,805	629	574	Fuel system rinsed with condensate No. 1 and rinse H <sub>2</sub> O in inner dump tanks; core dumped

Table 14. (Continued)

Date	Time	Activity (r/hr)			Remarks
		Fuel Cell	Reactor Cell	Heat Exchanger Cell	
3-4-54	3:55 PM	2,140	575	536	Rinsed fuel system No. 2, drained to outer dump tanks
3-5-54	8:35 AM	1,805	520	470	Condensate tanks full
3-5-54	1:30 PM	1,640	503	437	No. 3 rinse completed and system drained; condensate in weight tanks
3-8-54	8:15 AM	1,170	339	328	No. 4 rinse completed and system drained
3-8-54	4:30 PM	1,150	300	382	No. 5 rinse completed and ~30 kg in inner dump tanks
3-9-54	8:15 AM	1,090	344	388	Condensate weight tanks full
3-9-54	2:30 PM	1,040	274	328	No. 6 rinse completed
3-10-54	8:30 AM	1,200	274	328	Condensate in inner dump tanks
3-10-54	2:30 PM	875	219	273	Inner dump tanks full
3-11-54	8:10 AM	854	208	257	Heat exchanger empty; condensate in inner dump tanks
3-11-54	2:30 PM	766	186	224	Rinse No. 8 completed
3-12-54	9:00 AM	875	197	235	Inner dump tanks empty
3-12-54	4:00 PM	820	175	229	Condensate in inner dump tanks; rinse No. 9 completed
3-16-54	4:30 PM	710	148	142	160 kg of condensate in system
3-18-54	10:45 AM	690	132	125	160 kg of condensate drained to waste evaporator, system empty
3-18-54	4:30 PM	407	159	134	200 liters of 8 M HNO <sub>3</sub> in system
3-18-54	6:30 PM	306	123	115	HNO <sub>3</sub> drained to waste; system empty
3-19-54	2:30 PM	224	109	137	System rinsed and rinse drained to waste evaporator; system empty during measurement

Table 14. (Continued)

Date	Time	Activity (r/hr)			Remarks
		Fuel Cell	Reactor Cell	Heat Exchanger Cell	
3-20-54	2:30 PM	153	104	109	200 liters of alkaline tartrate in system
3-23-54	12:30 PM	213	104	109	Alkaline tartrate drained; fuel system empty
3-24-54	8:50 AM	192	98	115	System rinsed with water and drained
3-25-54	10:00 AM	137	93	93	System rinsed again with water and drained
3-29-54	12:15 PM	143	85	99	System rinsed with water again and drained
3-31-54	11:00 AM	279	49	58	5% HNO <sub>3</sub> circulated for ~ 9 hr and drained
3-31-54	3:30 PM	175	33	44	Rinsed with water and drained
4-1-54	1:00 PM	137	27	33	System rinsed with alkaline tartrate and drained
4-2-54	2:30 PM	98	27	38	Rinsed with water and drained
4-5-54	12:00 Noon	90	27	38	System rinsed with water and drained
4-7-54	4:00 PM	55	22	33	System completely filled with 8 M HNO <sub>3</sub> , circulated, and drained
4-8-54	5:30 PM	55	11	22	System completely filled with water and drained (461 liters)

<sup>a</sup>Probe in fuel cell at 19 ft 6 in. from roof of cell, in reactor and heat exchanger cells at 9 ft 6 in.

Table 15. Radiation Survey of Components in Storage Pool after Partial Chemical Decontamination and 90-day Decay

Component	Positions	Reading (r/hr)	Shielding
Main circulating pump	Intake flange	2	4-5/8 in. of air
	Discharge flange	2	4-5/8 in. of air
	Impeller-housing	1	At contact
	Rear	1	4-5/8 in. of air
Fuel feed pump	Discharge flange	0.60	At contact
	Filter	4.5	4-5/8 in. of air
	Check housing	5	At contact
High-pressure gas bleed valve	Low-pressure flange	0.80	At contact
	High-pressure flange	0.72	At contact
	Top of operator	0.04	At contact
Steamer pot, fed by condensate from dump-tank steam	Top of pot	2	At contact
	Middle of pot	2.8	At contact
	Bottom of pot	0.80	At contact
Off-gas condenser, vapors from catalytic recombiner	Top of condenser	1	At contact
	Middle of condenser	0.60	At contact
	Bottom of condenser	0.70	At contact
Steamer pot, fed by condensate from flame recombiner	Top of pot	1	At contact
	Middle of pot	1	At contact
	Bottom of pot	1	At contact
Gas condenser, for steam from dump-tank evaporator	Top (near the leak)	5.2	At contact
	Middle of condenser	1.2	At contact
	Bottom of condenser	1.4	At contact
Fuel dump valve and attached lines	Rupture-disk flange	3	At contact
	Open lines on rupture disk	2.2	At contact
	Low-pressure flange	3	At contact
	Top of operator	1	At contact
	High-pressure flange	1	At contact
	Valve seat	4	At contact
Flame recombiner	At spark plugs	6	At contact
	Middle of recombiner	0.30	At contact
	Bottom of recombiner	1.5	At contact
Core-to-dump-tank let-down heat exchanger	Top line	3	Through 10 in. of water
Core-to-dump-tank throttling valve	Top of operator	0.30	At contact
	Middle of valve	0.90	At contact
	Valve seat	3	At contact
	Low-pressure flange	0.30	At contact
Main-heat-exchanger tube bundle	At U section of tubes	2.4	Through 13.5 in. of water