

OAK RIDGE NATIONAL LABORATORY

OPERATED BY
UNION CARBIDE CORPORATION
NUCLEAR DIVISION



POST OFFICE BOX X
OAK RIDGE, TENNESSEE 37830

INTERNAL USE ONLY

ORNL
CENTRAL FILES NUMBER

68-3-38

DATE: March 25, 1968

SUBJECT: Decay Heat Generation by Fission Products and
 ^{233}Pa in a Single-Region Molten Salt Reactor

TO: M. W. Rosenthal and E. S. Bettis

FROM: W. L. Carter

COPY NO.

ABSTRACT

Fission product and ^{233}Pa decay heat and concentrations have been calculated for a single-region MSR for reactor equilibrium conditions and as a function of decay cooling time. The MSR is a 2000-Mw(e) system containing 2000 ft³ of LiF-BeF₂-ThF₄- $^{233}\text{UF}_4$ fuel. Three operating modes were studied: (1) inert gas sparging to remove noble gases from the fuel, (2) inert gas sparging plus removal of noble metals by reaction with surfaces of the heat exchanger loop, and (3) removal of all fission products by chemical processing only. In all three cases the fuel was being processed in a chemical plant on a 38-day cycle. Tabular and graphic data are presented for 32 fission product elements and ^{233}Pa for decay times up to 11 years.

NOTICE This document contains information of a preliminary nature and was prepared primarily for internal use at the Oak Ridge National Laboratory. It is subject to revision or correction and therefore does not represent a final report. The information is only for official use and no release to the public shall be made without the approval of the Legal and Information Control Department of Union Carbide Corporation, Nuclear Division.

INTRODUCTION

A primary concern in the design of a nuclear reactor is the removal of the after-heat when the reactor is shut down. The problem becomes acute when it is assumed that the shutdown is unscheduled due to an emergency that has disrupted the normal cooling system of the reactor. In the case of the Molten Salt Reactor, this requires draining the fuel into a receiver where emergency cooling is provided. Proper design of this emergency cooling system is therefore essential to safe operation of the reactor.

This study has been carried out to determine the afterheat as a function of time after fuel has been drained from the reactor. The reactor system considered is a 2000-Mw(e), single-region MSR containing 2000 ft³ of LiF-BeF₂-ThF₄-²³³UF₄ fuel; the composition of the fuel carrier is respectively 52, 36, 12 mole %. In addition to sufficient ²³³U for criticality, the fuel contains about 0.256 g/liter of ²³³Pa plus fission products. It was assumed that the reactor had been operating long enough so that fission products were present in equilibrium concentrations for the chosen processing conditions. Three processing modes were considered in determining fission product concentrations: (1) all fission products removed on a 38-day cycle through the processing plant; (2) noble gases removed by sparging and the remaining fission products removed by chemical processing; and (3) noble gases sparged, noble metals plated out on reactor surfaces, and the remaining nuclides removed by chemical processing.

Protactinium is removed on a three-day cycle in a separate processing step. About 7% of the ²³³Pa (14.5 kg) is in the circulating fuel; the remainder (190.5 kg) is held in the processing plant. The system has a breeding ratio of 1.076.

Heat generation and inventory data were calculated for both fission products and ²³³Pa from equilibrium to about 11 year's decay. At equilibrium gross fission products and ²³³Pa in the fuel are generating about 289.4 Mw and 0.74 Mw, respectively; an additional 9.7 Mw is being generated by ²³³Pa in the processing plant. When gases are sparged in the reactor, the fission product contribution is reduced to 257 Mw, and, when both noble gases and noble metals are absent, the rate is further reduced to 255.8 Mw. The fission product concentrations for these three processing modes are respectively 3.08, 1.94, and 1.48 g/liter.

The principal source of heat is the short-lived fission products having half-lives less than a few minutes. Hence, there is an initial large decrease in the heat generation rate when fissioning ceases. In the first 2 min after shutdown, the rate is down by a factor of 3, in 18 min by a factor of 4.5, in 1 hr by a factor of 7, and in 10 hr by a factor of 17. Protactinium-233, which has a 27.4-day half-life, does not show a significant decrease in heat production until about 15 to 20 days after shutdown.

Extensive tables and graphs in later sections of this memorandum describe more completely the thermal characteristics of this fuel for cooling times up to about 11 years.

A further interesting result of this study is the importance of just a few fission products in the total heat generation rate. For example, at equilibrium, Rb, Cs, and Sb, respectively, account for 22.8, 21, and 17.3% of fission product decay heat. At 1 hour's decay, the figures are 18.6% I, 13% Kr, 10.5% La, and 9.6% Y; at 10 hour's decay, the values are 23.8% I, 16.5% La, and 11.4% Y. For much longer decay times (e.g., 125 days), about 80% of the decay heat is due to Nb, Zr, Pr, and Y.

In the period two days to five months after reactor shutdown, more heat is being generated by the massive amount of ^{233}Pa in the processing plant than by decay of gross fission products (see Fig. 2, page 13).

METHOD OF CALCULATION

Equilibrium concentrations of fission products were calculated by the HTGN code written by Watson.¹ This program treats 290 fission product nuclides and accounts for their removal by chemical processing, neutron capture, decay, gas sparging, and sorption on reactor surfaces. Production is a function of the fission rate and characteristic yield. Decay heat and concentrations at equilibrium and after shutdown were calculated by the CALDRON code written by Carter.² This program treats 469 nuclides and was written to describe the behavior of fuel in a chemical processing plant. Beta heat, gamma heat, and concentration are calculated for each of the 469 nuclides as a function of time. The program accounts for branching in the decay chains, and, in the case of chemical processing, allows the removal and accumulation of specified nuclides in various process operations.

CHARACTERISTICS OF THE REACTOR

The molten salt reactor for which these calculations were made has the following characteristics:³

Fuel	LiF-BeF ₂ -ThF ₄ - ²³³ UF ₄
Composition of carrier salt, mole %	52-36-12
Power, Mw(th)	4444
Fuel volume in core, ft ³	1333
Total circulating fuel volume, ft ³	2000
Processing cycle time for fission products, days	38
Processing cycle time for ²³³ Pa, days	3
²³³ Pa inventory in circulating fuel, kg	14.5
²³³ Pa inventory in processing plant, kg	190.5
Breeding ratio	1.076

AVERAGE LIFETIMES OF NOBLE GASES AND NOBLE METALS

It is well established that for good breeding performance the fission product xenon must be quickly removed from the fission zone. This is accomplished by sparging the circulating fuel with an inert gas; this action also removes krypton. Competing with sparging for removal of noble gas atoms are radioactive decay, neutron capture, diffusion into the graphite moderator, and chemical processing. Studies and experience in MSRE operation indicate that the sparging rate needs to be sufficiently vigorous that the average residence time of a gas atom in the fuel is only about 50 sec for maintaining tolerable xenon poisoning.

Secondly, there is a group of fission products (Se, Nb, Mo, Tc, Ru, Rh, Pd, Ag, In, Te) whose behavior in the system is not entirely understood, but it is believed that these elements distribute throughout the circulating fuel loop by reacting with or otherwise attaching themselves to surfaces contacted by the fuel. This group is known as the noble metals. Competing events for the removal of the noble metals are radioactive decay, neutron capture, and chemical processing. The average lifetime for this "plating out" effect is probably different for each of these elements, and the data are not available for determining the values very accurately. MSRE data for fission product distribution in the reactor system were examined by Watson,⁴ who concluded that a value of 50 hr is reasonable for the average lifetime of the noble metals. The scatter and paucity of the data did not warrant assigning a characteristic lifetime to each element; so the 50-hr figure was assumed to apply to all.

REACTOR OPERATING CONDITIONS FOR WHICH DECAY HEAT RATES ARE COMPUTED

Fission Products

The three following situations were considered in the HTGN and CALDRON computations for determining the equilibrium heat and afterheat rates for decaying fission products:

1. Gross amounts of fission products in the fuel, that is, no sparging of noble gases and no plating out of noble metals.
2. Noble gases sparged on 50-sec cycle but no plating out of noble metals.
3. Noble gases sparged on 50-sec cycle and noble metals plated out on 50-hr cycle.

In each case equilibrium concentrations were calculated for a 38-day chemical processing cycle and the characteristic losses due to neutron absorption and radioactive decay. However, in the case of the gases, the computer program does not provide for removal due to diffusion into the graphite.

In the computer program treatment of Cases 2 and 3, sparging and plating out has the effect not only of removing the noble gases and noble metals but also of removing daughter products of these elements. While this treatment is quite proper for gases which are quantitatively removed from the fuel environment, it is not as rigorous for noble metal decay products. Noble metals attached to reactor, piping, and heat exchanger surfaces are always in contact with fuel, and decay products, which are not noble to these surfaces, might reenter the fuel stream. The calculations on Cases 2 and 3 include only the daughter products of the noble gases and metals that are associated with the equilibrium amounts of these gases and metals in the fuel.

Protactinium

The amount of ^{233}Pa present in the system had been determined previously by Kerr.⁵ His results stated that for a 3-day processing cycle for protactinium there would be 14.5 kg ^{233}Pa (0.256 g/liter) in the circulating fuel stream and an additional 190.5 kg in the processing plant. The calculation of ^{233}Pa afterheat was then straightforward, since the chain terminates with a single decay. The beta and gamma decay energies are respectively 9.3 and 41.5 w/g, totaling 50.8 w/g.

DISCUSSION OF RESULTS

Comparison of Cases 1, 2, and 3

The heat generation rates as a function of time after reactor shutdown given in Tables 1, 2, and 3 were calculated for the three assumed reactor operating conditions described above. A graphic presentation of the total $\beta + \gamma$ heat generation is given in Fig. 1. Values for ^{233}Pa in these exhibits are for ^{233}Pa in the fuel stream only.

The effect on decay heat rate of removing noble gases and noble metals is shown by the three fission product curves of Fig. 1. At equilibrium the effect of sparging and plating is to decrease the gross decay heat by about 11.6%. However, the effect on heat generation after fuel is dumped from the reactor is more pronounced, particularly during the first hour or so. During this period, heat generation for the sparged and "plated out" fuel is as much as 33% smaller than the gross fission product case (Table 4). After the first hour of decay, the effect of removing gases and noble metals is less pronounced but still reduces the decay heat by about 20% on the average for the next year. After three year's decay there is a considerably larger difference between the decay curves because the long-lived daughters of krypton and xenon are absent from the sparged fuel. However, by this time the decay heat generation rate is small even for gross fission products; so the significance of this difference is minor.

Table 1. Heat Generation from Fission Products and ^{233}Pa in Fuel of One-Region Molten Salt Reactor
With No Sparging of Noble Gases and No Plating of Noble Metals

Reactor Power = 4444 Mw(th)
 Fuel Volume in Reactor Circulating System = 2000 ft³
 Fuel Processing Cycle Time = 38 days
 ^{233}Pa Processing Cycle Time = 3 days
 Equilibrium ^{233}Pa Concentration = 7.25 g/ft³
 Equilibrium Fission Product Concentration = 87.16 g/ft³

Time After Fuel Dumped From Reactor (hr)		Fission Products in Fuel Stream			^{233}Pa in Fuel Stream		^{233}Pa + Fission Products
		β Heat (w/ft ³)	γ Heat (w/ft ³)	$\beta + \gamma$ Heat (w/ft ³)	$\beta + \gamma$ Heat (w/ft ³)	(g/ft ³)	$\beta + \gamma$ Heat (w/ft ³)
0	(equilibrium)	0.1194×10^6	0.2531×10^5	0.1447×10^6	368.3	7.250	0.1451×10^6
0.001	(3.6 sec)	0.7109×10^5	0.2526×10^5	0.9635×10^5	368.3	7.250	0.9672×10^5
0.003	(10.8 sec)	0.4577×10^5	0.2505×10^5	0.7082×10^5	368.3	7.250	0.7119×10^5
0.01	(36 sec)	0.3309×10^5	0.2433×10^5	0.5742×10^5	368.3	7.250	0.5779×10^5
0.03	(1.8 min)	0.2686×10^5	0.2291×10^5	0.4977×10^5	368.3	7.250	0.5014×10^5
0.10	(6 min)	0.2054×10^5	0.2062×10^5	0.4116×10^5	368.2	7.249	0.4153×10^5
0.30	(18 min)	0.1453×10^5	0.1729×10^5	0.3182×10^5	368.2	7.248	0.3219×10^5
1.0		0.8940×10^4	0.1240×10^5	0.2134×10^5	367.9	7.242	0.2171×10^5
3.0		0.5917×10^4	0.8398×10^4	0.1432×10^5	367.1	7.227	0.1469×10^5
10		0.3464×10^4	0.5127×10^4	0.8591×10^4	364.4	7.174	0.8955×10^4
30		0.1945×10^4	0.3356×10^4	0.5300×10^4	356.8	7.024	0.5657×10^4
100	(4.17 days)	0.1119×10^4	0.2090×10^4	0.3209×10^4	331.5	6.525	0.3540×10^4
300	(12.5 days)	0.6762×10^3	0.1132×10^4	0.1809×10^4	268.5	5.285	0.2078×10^4
1,000	(41.7 days)	0.2933×10^3	0.3781×10^3	0.6714×10^3	128.4	2.528	0.7998×10^3
3,000	(125 days)	0.1021×10^3	0.1161×10^3	0.2182×10^3	15.6	0.3071	0.2338×10^3
10,000	(1.14 years)	0.2203×10^2	0.9287×10^1	0.3131×10^2	0.01	0.000192	0.3132×10^2
30,000	(3.42 years)	0.4463×10^1	0.1664×10^1	0.6127×10^1			0.6127×10^1
100,000	(11.4 years)	0.1556×10^1	0.8611×10^0	0.2417×10^1			0.2417×10^1

Ref: Case JW-9

Table 2. Heat Generation From Fission Products and ^{233}Pa in Fuel of One-Region Molten Salt Reactor
With Sparging of Noble Gases but No Plating of Noble Metals

Cycle Time for Noble Gas Sparging = 50 sec
 Reactor Power = 4444 Mw(th)
 Fuel Volume in Reactor Circulating System = 2000 ft³
 Fuel Processing Cycle Time = 38 days
 ^{233}Pa Processing Cycle Time = 3 days
 Equilibrium ^{233}Pa Concentration = 7.25 g/ft³
 Equilibrium Fission Product Concentration = 54.85 g/ft³

Time After Fuel Dumped From Reactor (hr)		Fission Products in Fuel Stream			^{233}Pa in Fuel Stream		^{233}Pa + Fission Products
		β Heat (w/ft ³)	γ Heat (w/ft ³)	$\beta + \gamma$ Heat (w/ft ³)	$\beta + \gamma$ Heat (w/ft ³)	(g/ft ³)	$\beta + \gamma$ Heat (w/ft ³)
0	(equilibrium)	0.1114×10^6	0.1715×10^5	0.1285×10^6	368.3	7.250	0.1289×10^6
0.001	(3.6 sec)	0.6357×10^5	0.1710×10^5	0.8068×10^5	368.3	7.250	0.8105×10^5
0.003	(10.8 sec)	0.3884×10^5	0.1692×10^5	0.5576×10^5	368.3	7.250	0.5613×10^5
0.01	(36 sec)	0.2682×10^5	0.1626×10^5	0.4308×10^5	368.3	7.250	0.4345×10^5
0.03	(1.8 min)	0.2129×10^5	0.1499×10^5	0.3629×10^5	368.3	7.250	0.3666×10^5
0.10	(6 min)	0.1609×10^5	0.1316×10^5	0.2925×10^5	368.2	7.249	0.2962×10^5
0.30	(18 min)	0.1122×10^5	0.1096×10^5	0.2217×10^5	368.2	7.249	0.2254×10^5
1.0		0.6800×10^4	0.8362×10^4	0.1516×10^5	367.9	7.242	0.1553×10^5
3.0		0.4873×10^4	0.6650×10^4	0.1152×10^5	367.1	7.227	0.1189×10^5
10		0.3141×10^4	0.4809×10^4	0.7950×10^4	364.4	7.174	0.8314×10^4
30		0.1768×10^4	0.3270×10^4	0.5038×10^4	356.8	7.024	0.5395×10^4
100	(4.17 days)	0.9729×10^3	0.2034×10^4	0.3007×10^4	331.5	6.525	0.3338×10^4
300	(12.5 days)	0.5662×10^3	0.1103×10^4	0.1669×10^4	268.5	5.285	0.1937×10^4
1000	(41.7 days)	0.2286×10^3	0.3717×10^3	0.6003×10^3	128.4	2.528	0.7287×10^3
3000	(125 days)	0.8058×10^2	0.1141×10^3	0.1947×10^3	15.6	0.3071	0.2103×10^3
10,000	(1.14 years)	0.2051×10^2	0.7621×10^1	0.2814×10^2	0.01	0.000192	0.2815×10^2
30,000	(3.42 years)	0.3475×10^1	0.4481×10^0	0.3923×10^1			0.3923×10^1
100,000	(11.4 years)	0.7705	0.1218	0.8923			0.8923

Ref: Case JW-9R

Table 3. Heat Generation from Fission Products and ^{233}Pa in Fuel of One-Region Molten Salt Reactor
With Sparging of Noble Gases and Plating of Noble Metals

Cycle Time for Sparging of Noble Gases = 50 sec
 Cycle Time for Plating of Noble Metals = 50 hr
 Reactor Power = 4444 Mw(th)
 Fuel Volume in Reactor Circulating System = 2000 ft³
 Fuel Processing Cycle Time = 38 days
 ^{233}Pa Processing Cycle Time = 3 days
 Equilibrium ^{233}Pa Concentration = 7.25 g/ft³
 Equilibrium Fission Product Concentration = 41.85 g/ft³

Time After Fuel Dumped From Reactor (hr)		Fission Products in Fuel Stream			^{233}Pa in Fuel Stream		^{233}Pa + Fission Products
		β Heat (w/ft ³)	γ Heat (w/ft ³)	$\beta + \gamma$ Heat (w/ft ³)	$\beta + \gamma$ Heat (w/ft ³)	^{233}Pa (g/ft ³)	$\beta + \gamma$ Heat (w/ft ³)
0	(equilibrium)	0.1113×10^6	0.1658×10^5	0.1279×10^6	368.3	7.250	0.1283×10^6
0.001	(3.6 sec)	0.6348×10^5	0.1654×10^5	0.8002×10^5	368.3	7.250	0.8039×10^5
0.003	(10.8 sec)	0.3875×10^5	0.1635×10^5	0.5510×10^5	368.3	7.250	0.5547×10^5
0.01	(36 sec)	0.2624×10^5	0.1570×10^5	0.4193×10^5	368.3	7.250	0.4230×10^5
0.03	(1.8 min)	0.2122×10^5	0.1443×10^5	0.3565×10^5	368.3	7.250	0.3602×10^5
0.10	(6 min)	0.1601×10^5	0.1258×10^5	0.2859×10^5	368.2	7.249	0.2896×10^5
0.30	(18 min)	0.1111×10^5	0.1034×10^5	0.2145×10^5	368.2	7.249	0.2182×10^5
1.0		0.6638×10^4	0.7654×10^4	0.1429×10^5	367.9	7.242	0.1466×10^5
3.0		0.4631×10^4	0.5821×10^4	0.1045×10^5	367.1	7.227	0.1082×10^5
10		0.2880×10^4	0.3963×10^4	0.6843×10^4	364.4	7.174	0.7207×10^4
30		0.1552×10^4	0.2543×10^4	0.4095×10^4	356.8	7.024	0.4452×10^4
100	(4.17 days)	0.8577×10^3	0.1610×10^4	0.2468×10^4	331.5	6.525	0.2800×10^4
300	(12.5 days)	0.5410×10^3	0.9729×10^3	0.1514×10^4	268.5	5.285	0.1782×10^4
1000	(41.7 days)	0.2226×10^3	0.3285×10^3	0.5511×10^3	128.4	2.528	0.6797×10^3
3000	(125 days)	0.7826×10^2	0.1050×10^3	0.1833×10^3	15.6	0.3071	0.1989×10^3
10,000	(1.14 years)	0.1971×10^2	0.7514×10^1	0.2722×10^2	0.01	0.000192	0.2723×10^2
30,000	(3.42 years)	0.3310×10^1	0.4346	0.3745×10^1			0.3745×10^1
100,000	(11.4 years)	0.7698	0.1218	0.8916			0.8916

Ref: Case JW-9RP

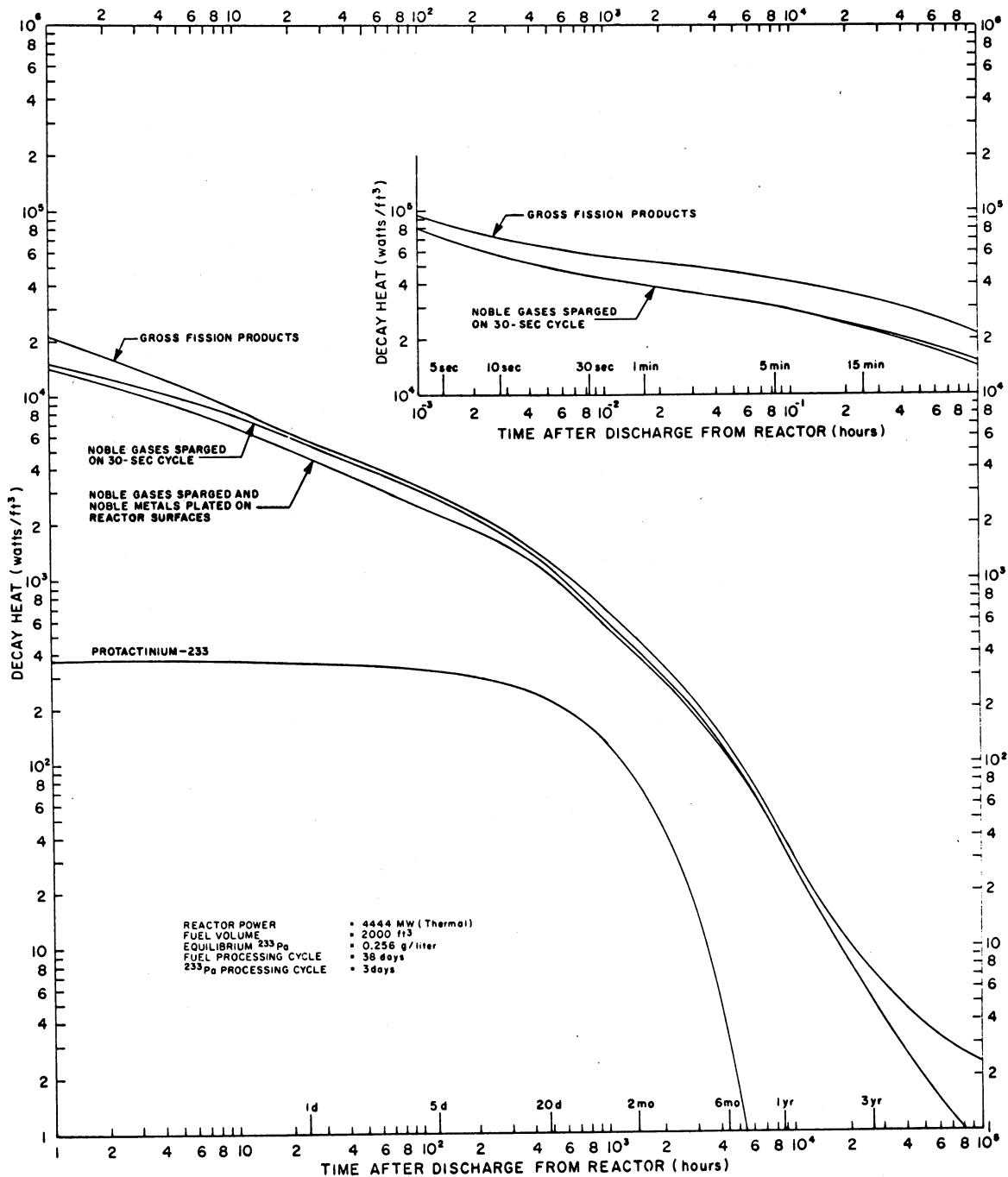


Fig. 1. Fission Product and Protactinium Decay Heat in One-Region MSR Fuel.

Table 4. Relative Decrease in Fission Product Decay Heat Generation Rate When Noble Gases Are Sparged and Noble Metals Are Plated Out in Reactor

Time After Reactor Shutdown (hr)	Percent of Gross Heat Generation	
	Noble Gases Sparged ^a	Noble Gases Sparged Plus Noble Metals Plated Out ^a
0 (equilibrium)	88.8	88.4
0.001 (3.6 sec)	83.7	83.1
0.003 (10.8 sec)	78.7	77.8
0.01 (36 sec)	75	73
0.03 (1.8 min)	72.9	71.6
0.10 (6 min)	71.1	69.5
0.30 (18 min)	69.7	67.4
1.0	71	67
3.0	80.4	73
10	92.5	80.5
30	95	77.3
100 (4.17 days)	93.7	77
300 (12.5 days)	92.3	83.7
1,000 (41.7 days)	89.4	82.1
3,000 (125 days)	89.2	84
10,000 (1.14 years)	89.9	86.9
30,000 (3.42 years)	64	61.1
100,000 (11.4 years)	36.9	36.9

^a Also includes the decay heat of daughters of removed gases or noble metals.