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THE AMOUNTS OF FISSION PRODUCT NUCLIDES PRODUCED IN ^{239}PU-FUELLED FAST REACTORS AND THE RELATED HEAT GENERATION AFTER SHUT-DOWN

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1. INTRODUCTION

To obtain the decay heat generated by the fission products of ^{239}Pu, first the concentration of individual fission products was calculated as a function of in-pile time, cooling time, and specific power of the reactor. The equations used and the results of these calculations are presented and discussed in chapter 2. On the basis of these results, the activities and the decay power of individual fission product nuclides was calculated and, finally, these quantities were summed up to result in data for gross fission products. These latter calculations and their results are presented and discussed in chapter 3.

2. CONCENTRATION OF FISSION PRODUCT NUCLIDES

2.1 Basis of calculation

The concentrations of fission product nuclides have been calculated according to the following equation:

$$(1)\quad N_i\,(t,T) = 3{,}12\cdot 10^{10}\cdot B \left\{ y_i \frac{1-e^{-(\lambda_i+\sigma_i\ \phi)t}}{\lambda_i+\sigma_i\ \phi} + \frac{y_{Mi}\ \sigma_{Mi}\ \phi_i}{\lambda_{Mi} + \sigma_{Mi}\phi} \quad x \right.$$

$$\left. \left(\frac{1-e^{-(\lambda_i+\sigma_i\ \phi)t}}{\lambda_i+\sigma_i\ \phi} - \frac{e^{-(\lambda_{Mi}+\sigma_{Mi}\ \phi)t} - e^{-(\lambda_i+\sigma_i\ \phi)t}}{\lambda_i-\lambda_{Mi}+(\sigma_i-\sigma_{Mi})\ \phi} \right) \right\} e^{-\lambda_i T}$$

where

$N_i(t,T)$	$[\mathrm{cm}^{-3}]$	concentration of fission product nuclide i	
t	$[\mathrm{sec}]$	in-pile time	
T	$[\mathrm{sec}]$	cooling time	
B	$[\frac{\mathrm{kW}}{\mathrm{l}}]$	specific power of the reactor	
y_i		yield	
λ_i	$[\mathrm{sec}^{-1}]$	decay constant	of fission product nuclide i
σ_i	$[\mathrm{cm}^2]$	(n, γ) cross section	
ϕ	$[\mathrm{cm}^{-2}\mathrm{sec}^{-1}]$	neutron flux, summed over all energies and averaged over the core volume	

Subscript M	Quantity refers to nuclide, that is transformed to nuclide i by (n,γ) process

For a nuclide with half life of less than one year, $\sigma_i\phi$ was neglected as compared to λ_i and, in addition, the generation of the nuclide by (n, γ) capture of another nuclide was neglected. Nuclides with half lives less than 5 days were not considered. The yields for the short lived nuclides were taken to be equal to those for the stable or long lived nuclides that form the end products of the respective chains. The numerical values for yields and half-lives that were used for the calculations are shown in table 1, the yields were taken from tables given in [1]. The (n,γ) cross sections were taken from [2], [3] and [4] as a function of energy. To obtain the one-group cross sections that enter in (1), the multi-group cross sections were weighed with a neutron spectrum averaged over the core volume of the reactor in question. This spectrum was calculated by one dimensional diffusion theory in 26 energy groups by means of the Karlsruhe nuclear code system NUSYS.

The number of fission product nuclides within a kg of fuel is obtained as

$$(2) \quad n_i\ (t,\ T) = \frac{10^3 N_i(t,T)}{\rho \cdot \omega}$$

and the nuclide concentrations are converted to weights by

$$(3) \quad F_i = \frac{10^3 \cdot N_i(t,T) \cdot A_i}{L\ \rho\ \omega}$$

where

F_i [$\frac{g}{\text{kg fresh fuel}}$]	total weight of fission product nuclide i relative to 1 kg of fresh fuel
A_i	atomic weight of fission product nuclide i
L	Avogadro's number
ρ	fuel fraction in core
ω [$g\ cm^{-3}$]	fuel density

2.2 Results of calculations

The amounts of fission products available after various cooling times were calculated for the sodium cooled fast breeder reactor design Na-1 [5] and for the steam cooled fast breeder reactor design D-1 [6]. The relevant reactor data are shown in table 2. Tables 3 and 4 show the related amounts of fission product nuclides for various cooling times and of fission product chemical elements for a cooling time of 100 days.

3. ACTIVITY OF FISSION PRODUCT NUCLIDES AND PRODUCTION OF DECAY HEAT

3.1 Basis of calculation

The heat generated after reactor shut-down is obtained by summing up the fractions of the decay energies absorbed in the reactor vessel for all radioactive nuclides present. In average, half of the decay energy is carried away by the neutrinos and, therefore, will not be transformed into heat. The energy due to β- and γ-radiation, on the other hand, will be totally absorbed.

The activity of 1 kg fuel after a cooling time T is given by

$$(4) \qquad Z\,(t,T)\left[\frac{\text{Curie}}{\text{kg}}\right] = \sum_i \frac{\lambda_i\, n_i\,(t,T)}{3.7 \cdot 10^{10}}$$

The corresponding heat production by β- and γ-radiation amounts to:

$$(5) \qquad P_\beta\,(t,T)\left[\frac{\text{W}}{\text{kg}}\right] = \sum_i E_{\beta_i}\, \lambda_i\, n_i(t,T)$$

and

$$(6) \qquad P_\gamma\,(t,T)\left[\frac{\text{W}}{\text{kg}}\right] = \sum_i E_{\gamma i}\, \lambda_i n_i(t,T).$$

The total heat production is given by the sum of the β- and γ-contributions:

$$(7) \qquad P\,(t,T) = P_\beta\,(t,T) + P_\gamma(t,T)$$

where

$$\left.\begin{array}{lll} E_{\beta i} & [\text{Wsec}] & \text{mean energy of } \beta\text{-radiation} \\ E_{\gamma i} & [\text{Wsec}] & \text{mean energy of } \gamma\text{-radiation} \end{array}\right\} \text{ of nuclide i}$$

Values for the mean energies were taken from the tables by King and Perkins /7/. The more important data have been checked using recently edited tables /8/ /9/.

3.2 Discussion of the influence of approximations made

The error due to the neglection of fission products with half lives of less than 5 days was found to be less than 1 % for cooling times $t > 30$ d. The heat contribution due to the higher isotopes of uranium and plutonium does not exceed 1 % of the total effect and, therefore, was also neglected.

Eight nuclides considered have daughter nuclides with half lives of less than 5 days. These can be taken into account with sufficient accuracy, if the decay energy of the mother nuclide is replaced by

$$E_m + \frac{\lambda_d}{\lambda_d - \lambda_m} E_d ,$$

where the subscripts m and d refer to mother and daughter nuclide. The factor of E_d was found to deviate appreciably from unity for the transition $^{140}\mathrm{Ba} \longrightarrow {}^{140}\mathrm{La}$ only.

3.3 Results of calculations

Tables 5 and 6 present numerical values of the specific activities and of the corresponding heat production for all nuclides considered in this paper for the reactor designs Na-1 and D-1. These data are given for three cooling times. Figures 1 to 3 show the expressions just mentioned after they have been summed over all the nuclides as a function of cooling time, the numerical values for the total heat production generated by β- and γ-radiation are also shown in table 7. An error of $\pm$ 5 % is assigned to these data, due to errors in input data and to approximations made.
The time behaviour of the heat production as a function of in pile-time t and cooling time T is usually approximated by a Way Wigner formula

$$(8) \qquad P(t,T) = K \left[T^{-x} - (T+t)^{-x} \right].$$

This formula expresses the results obtained in this report quite well, if the following values are chosen for the free parameters

$$x = 0.33$$

$$K = 855 \ \frac{W}{kg} \quad \text{for Na-1}$$

$$K = 639 \ \frac{W}{kg} \quad \text{for D-1}$$

Here, t and T have to be measured in days. It was checked, that the exact results are approximated to ± 3 % for T = 6oo d if these parameters are used in (8). It is possible, to express the time behaviour of the heat production by fission products for Na-1 and D-1 fuel (s. table 7) by a single formula (8) with $x = 0.33$ and $K = 6.45 \; 10^3 \; \frac{W}{MW_{th}}$ x). Apparently, the difference in neutron spectrum of the two reactor types is insignificant in this respect.

Fig. 4 shows the heat production for various in-pile times as a function of cooling time. The curve related to $t = \infty$ was calculated by means of equation (8). The other curves were obtained by a more exact procedure: The time dependence of the contributions of ^{106}Ru and ^{144}Ce was calculated explicitely (according to $1-e^{-\lambda t}$),the remaining nuclides were divided into two groups with average half life of 50 d and 15 a. In addition, Fig. 4 shows curves for $t = \infty$ for ^{235}U taken from [7] and [10]. From these two sets of curves, those due to King and Perkins are considered more reliable [11]. Calorimetric measurements made by Johnston [12] in 1965 on ^{239}Pu after a in-pile time of 37 days resulted in a decay power by 6 % less than for ^{235}U. The higher values for the longer irradiation times considered in this report are mainly due to the higher yield of long lived ^{106}Ru that accompanies the fission of ^{239}Pu.

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x) In case P is expressed in units $\frac{W}{MW_{th}}$. Note the relation

$$P \left[\frac{W}{MW_{th}} \right] = \frac{t}{am} \; P \left[\frac{W}{kg} \right]$$

t in-pile time (days)
am burnup $\left[\frac{MWd}{kg} \right]$

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TABLE 1. DATA OF FISSION PRODUCT NUCLIDES

	Half life	$\bar{E}_{\beta}$ [MeV]	$\bar{E}_{\gamma}$ [MeV]	Cumulative yield
Kr 85	10.6 a	0.221	0.004	$5.40 \cdot 10^{-3}$ (Rb)
Sr 89	50.4 d	0.556	0.000	$1.71 \cdot 10^{-2}$ (Y)
Sr 90 + Y 90	28 a	1.081	0.000	$2.24 \cdot 10^{-2}$ (Zr)
Y 91	58 d	0.593	0.004	$2.60 \cdot 10^{-2}$ (Zr)
Zr 95 + Nb 95^{m}	65 d	0.111	0.974	$5.00 \cdot 10^{-2}$ (Mo)
Nb 95	35 d	0.045	0.760	$5.00 \cdot 10^{-2}$ (Mo)
Ru 103 + Rh 103^{m}	40 d	0.104	0.499	$5.65 \cdot 10^{-2}$ (Rh)
Ru 106 + Rh 106	1 a	1.368	0.328	$4.70 \cdot 10^{-2}$ (Pd)
Cd 113^{m}	14 a	0.195	0.270	$6.50 \cdot 10^{-2}$ (Cd)
Cd 115^{m}	43 d	0.595	0.030	$3.50 \cdot 10^{-4}$ (In)
Te 129^{m} + Te 129	33 d	0.563	0.236	$1.40 \cdot 10^{-2}$ (I)
I 131	8.0 d	0.183	0.392	$3.79 \cdot 10^{-2}$ (Xe)
Xe 133	5.3 d	0.155	0.027	$6.90 \cdot 10^{-2}$ (Cs)
Cs 137 + Ba 137^{m}	30 a	0.239	0.595	$6.30 \cdot 10^{-2}$ (Ba)
Ba 140 + La 140	12.8 d	0.799	2.962	$5.56 \cdot 10^{-2}$ (Ce)
Ce 141	32.5 d	0.146	0.097	$5.30 \cdot 10^{-2}$ (Pr)
Ce 144 + Pr 144	277 d	1.288	0.080	$3.84 \cdot 10^{-2}$ (Nd)
Pr 143	13.7 d	0.315	0.000	$4.57 \cdot 10^{-2}$ (Nd)
Nd 147	11.1 d	0.271	0.137	$1.94 \cdot 10^{-2}$ (Sm)
Pm 147	2.65a	0.062	0.000	$1.94 \cdot 10^{-2}$ (Sm)
Sm 151	93 a	0.023	0.000	$7.80 \cdot 10^{-3}$ (Eu)
Eu 154	16 a	0.233	1.160	(n,γ) process of Eu 153
Eu 155	1.7 a	0.040	0.090	$2.10 \cdot 10^{-3}$ (Gd)
Eu 156	15 d	0.420	1.160	$1.10 \cdot 10^{-3}$ (Gd)

TABLE 2. REACTOR DATA

	Na-1	D-1
Specific power B $[\frac{kW}{l}]$	380	400
Fuel fraction ω	0.305	0.43
Fuel density[x)] ρ $[\frac{g}{cm^3}]$	9.4	9.4
Burn up $[\frac{MWd}{t}]$	80,000	55,000
In-pile time $[d]$	604	556

[x)] density of the oxide

TABLE 3. AMOUNTS OF FISSION PRODUCTS IN G/KG FRESH FUEL, FROM NA-1 AND D-1 FAST BREEDER REACTOR TYPES

Z	Element	A	Na-1 cooling time [days]					D-1 cooling time [days]					β, γ Activity [MeV]
			200	150	100	50	30	200	150	100	50	30	
35	Br	81	0.0503	0.0503	0.0503	0.0503	0.0503	0.0346	0.0346	0.0346	0.0346	0.0346	
34	Se	82	0.0063	0.0063	0.0063	0.0063	0.0063	0.0019	0.0019	0.0019	0.0019	0.0019	
36	Kr	85	0.1503	0.1517	0.1531	0.1545	0.1550	0.1038	0.1047	0.1057	0.1066	0.1070	β:0.83;0.67 γ:0.31;0.15
	Kr	83,84,86	0.4609	0.4609	0.4609	0.4609	0.4609	0.3181	0.3181	0.3181	0.3181	0.3181	
	Kr	total	0.6112	0.6126	0.6140	0.6154	0.6159	0.4219	0.4228	0.4238	0.4247	0.4251	
37	Rb	total (85+87)	0.4559	0.4559	0.4559	0.4559	0.4559	0.3115	0.3115	0.3115	0.3115	0.3115	
38	Sr	89	0.0042	0.0083	0.0166	0.0329	0.0434	0.0031	0.0062	0.0124	0.0246	0.0324	β:1.46
	Sr	90	0.6982	0.7006	0.7029	0.7053	0.7063	0.4806	0.4823	0.4839	0.4856	0.4862	β:1.54
	Sr	88	0.4476	0.4476	0.4476	0.4476	0.4476	0.3079	0.3079	0.3079	0.3079	0.3079	
	Sr	total	1.1500	1.1565	1.1671	1.1858	1.1973	0.7916	0.7964	0.8042	0.8181	0.8265	
39	Y	91	0.0107	0.0195	0.0355	0.0646	0.0820	0.0080	0.0146	0.0265	0.0482	0.0612	β:1.55
	Y	89	0.5405	0.5364	0.5281	0.5118	0.5013	0.3717	0.3686	0.3624	0.3502	0.3424	
	Y	total	0.5512	0.5559	0.5636	0.5764	0.5833	0.3797	0.3822	0.3889	0.3984	0.4036	
40	Zr	95	0.0314	0.0533	0.0908	0.1548	0.1916	0.0233	0.0398	0.0678	0.1154	0.1429	β:0.4;0.36 γ:0.76;0.73
	Zr	90,91,92,93 94, 96	6.4851	6.4763	6.4603	6.4312	6.4138	4.4354	4.4288	4.4169	4.3952	4.3822	
	Zr	total	6.5164	6.5296	6.5511	6.5860	6.6054	4.4587	4.4686	4.4847	4.5106	4.5251	
41	Nb	95	0.0331	0.0543	0.0794	0.1197	0.1323	0.0245	0.0402	0.0591	0.0891	0.1055	β:0.16; γ:0.76;0.23

TABLE 4. AMOUNTS OF FISSION PRODUCT CHEMICAL ELEMENTS FROM NA-1 AND D-1 FAST BREEDER REACTOR TYPES AFTER 100 DAYS OF COOLING

Fission Product Chemical Elements $\left[\frac{g}{\text{kg fresh fuel}}\right]$	Reactortype	
	Na-1	D-1
Br	0,0503	0,0346
Se	0,0063	0,0019
Kr	0,6140	0,4238
Rb	0,4559	0,3115
Sr	1,1671	0,8042
Y	0,5636	0,3889
Zr	6,5511	4,4847
Nb	0,0794	0,0591
Mo	8,0063	5,4942
Tc	1,9043	1,3420
Ru	7,4618	5,1851
Rh	1,9855	1,3704
Pd	4,7026	3,1909
Ag	0,5343	0,3639
Cd	0,2274	0,1573
In	0,0135	0,0093
Te	1,3109	0,9700
I	0,7834	0,5386
Xe	11,0777	7,6272
Cs	9,5336	6,5267
Ba	3,3106	2,2853
La	2,8557	1,9375
Ce	6,1442	4,2870
Pr	2,6144	1,7874
Nd	7,9067	8,7237
Pm	0,7720	0,5392
Sm	1,8374	1,2754
Eu	0,2212	0,1977
Gd	0,1526	0,1110
Total	82,8438	60,4185

TABLE 5 . ACTIVITY Z [Curie/kg], HEAT PRODUCTION [Watt/kg] OF FISSION PRODUCTS PRESENT IN 1 kg OF Na-1 FUEL

Cooling Time:	30 d			100 d			200 d		
Nuclide	Z[C/kg]	P_β[W/kg]	P_γ[W/kg]	Z[C/kg]	P_β[W/kg]	P_γ[W/kg]	Z[C/kg]	P_β[W/kg]	P_γ[W/kg]
Kr 85	61.4	0.080	0.012	60.7	0.079	0.001	59.2	0.077	0.001
Sr 89	956.3	4.151	0.000	482.5	1.588	0.000	121.7	0.400	0.000
Sr 90	201.6	0.645	0.000	200.9	0.642	0.000	199.4	0.637	0.000
Y 91	2025.2	7.114	0.055	876.7	3.080	0.024	264.1	0.928	0.007
Zr 95	4119.4	2.651	23.288	1952.2	1.256	11.036	674.9	0.434	3.816
Nb 95	5238.7	1.381	23.520	3126.4	0.820	14.098	1302.2	0.344	5.879
Ru 103	7486.8	2.313	11.080	2225.8	0.688	3.294	391.9	0.121	0.580
Ru 106	6631.7	26.880	6.420	5805.4	23.530	5.620	4806.2	19.480	4.653
Cd 113	5.8	0.001	0.009	5.8	0.001	0.009	5.4	0.001	0.009
Cd 115	23.7	0.083	0.004	7.9	0.028	0.001	1.5	0.005	0.000
Te 129	1400.5	2.782	1.163	325.9	0.648	0.271	37.9	0.075	0.032
I 131	321.7	0.263	0.747	0.7	0.028	0.002	$1.3\cdot10^{-4}$	0.000	0.000
Xe 133	147.9	0.136	0.024	$1.6\cdot10^{-2}$	0.000	0.000	$3.3\cdot10^{-8}$	0.000	0.000
Cs 137	506.8	0.377	0.926	504.6	0.375	0.922	501.0	0.372	0.915
Ba 140	2638.0	5.820	21.508	62.6	0.133	0.491	0.32	0.001	0.003
Ce 141	3114.0	2.697	1.787	699.0	0.605	0.401	82.1	0.071	0.047
Ce 144	6490.9	21.636	1.567	5471.9	18.240	1.321	4256.2	14.187	1.027
Pr 143	1120.6	2.094	0.000	33.3	0.062	0.000	0.2	0.000	0.000
Nd 147	335.0	0.538	0.271	4.0	0.006	0.003	$8.0\cdot10^{-3}$	0.000	0.000
Pm 147	746.0	0.247	0.000	709.5	0.260	0.000	659.6	0.242	0.000
Sm 151	8.2	0.001	0.000	8.2	0.001	0.000	7.3	0.001	0.000
Eu 154	4.8	0.007	0.034	4.8	0.007	0.033	4.7	0.001	0.032
Eu 155	111.1	0.026	0.059	102.7	0.024	0.055	91.4	0.022	0.049
Eu 156	27.9	0.069	0.192	1.1	0.003	0.008	$1.4\cdot10^{-2}$	0.000	0.000
Sum:	44030	82.018	92.669	22672	52.076	37.591	13467	37.401	17.049

TABLE 6. ACTIVITY Z [Curie/kg], HEAT PRODUCTION [Watt/kg] OF FISSION PRODUCTS PRESENT IN 1 kg OF D-1 FUEL

Cooling Time:	50 d			100 d			200 d		
Nuclide	Z [C/kg]	P_β [W/kg]	P_γ [W/kg]	Z [C/kg]	P_β [W/kg]	P_γ [W/kg]	Z [C/kg]	P_β [W/kg]	P_γ [W/kg]
Kr 85	42.4	0.055	0.001	41.9	0.055	0.001	41.1	0.054	0.001
Sr 89	715.1	2.353	0.000	360.4	1.186	0.000	90.1	0.296	0.000
Sr 90	138.8	0.444	0.000	138.3	0.442	0.000	137.3	0.439	0.000
Y 91	1190.4	4.182	0.033	654.5	2.299	0.018	197.6	0.694	0.005
Zr 95	2481.1	1.597	14.026	1457.7	0.983	8.241	500.9	0.322	2.832
Nb 95	3501.7	0.929	15.837	2328.4	0.616	10.510	955.8	0.258	4.360
Ru 103	3952.1	1.221	5.849	1663.0	0.514	2.461	290.8	0.090	0.430
Ru 106	4556.2	18.467	4.411	4142.6	16.791	4.010	3426.3	13.887	3.317
Cd 113	4.0	0.000	0.006	4.0	0.000	0.006	3.9	0.000	0.006
Cd 115	13.2	0.046	0.002	5.3	0.019	0.001	1.2	0.004	0.000
Te 129	684.8	1.361	0.569	236.8	0.470	0.197	30.9	0.061	0.026
I 131	37.1	0.026	0.086	0.6	0.000	0.001	$4.3\cdot10^{-4}$	0.000	0.000
Xe 133	8.1	0.007	0.001	$1.4\cdot10^{-2}$	0.000	0.000	$2.8\cdot10^{-8}$	0.000	0.000
Cs 137	348.4	0.259	0.636	347.3	0.258	0.634	345.1	0.256	0.630
Ba 140	657.0	1.464	5.438	47.1	0.099	0.338	0.14	0.001	0.002
Ce 141	1517.2	1.314	0.871	393.4	0.453	0.300	62.5	0.054	0.038
Ce 144	4941.8	14.973	1.084	3975.6	13.252	0.960	3114.7	10.382	0.752
Pr 143	300.2	0.561	0.000	26.6	0.050	0.000	0.13	0.000	0.000
Nd 147	71.8	0.115	0.058	3.2	0.005	0.003	$1.9\cdot10^{-2}$	0.000	0.000
Pm 147	512.7	0.188	0.000	495.6	0.182	0.000	461.2	0.169	0.000
Sm 151	5.6	0.001	0.000	5.6	0.001	0.000	5.6	0.001	0.000
Eu 154	3.7	0.005	0.025	3.6	0.005	0.025	3.6	0.005	0.025
Eu 155	76.5	0.018	0.041	72.4	0.017	0.038	64.6	0.015	0.034
Eu 156	8.9	0.022	0.061	0.9	0.002	0.006	$6.8\cdot10^{-3}$	0.000	0.000
Sum:	31739	59.914	70.074	16404	37.658	27.789	9733	26.990	12.424

TABLE 7. TOTAL HEAT PRODUCTION OF FISSION PRODUCTS

Cooling time:		30 d	50 d	100 d	150 d	200 d
P /‾W/kg_7	Na-1	175	134	90	69	54
	D-1	130	99	66	50	39
P /‾W/MW$_{th}$_7	Na-1	1325	1015	680	520	410
	D-1	1310	1000	665	505	397

Fig. 1 Heat Production due to β - and γ - Decay for Reactor Na - 1

in pile time 604 d burnup 80 $\frac{MWd}{kg}$

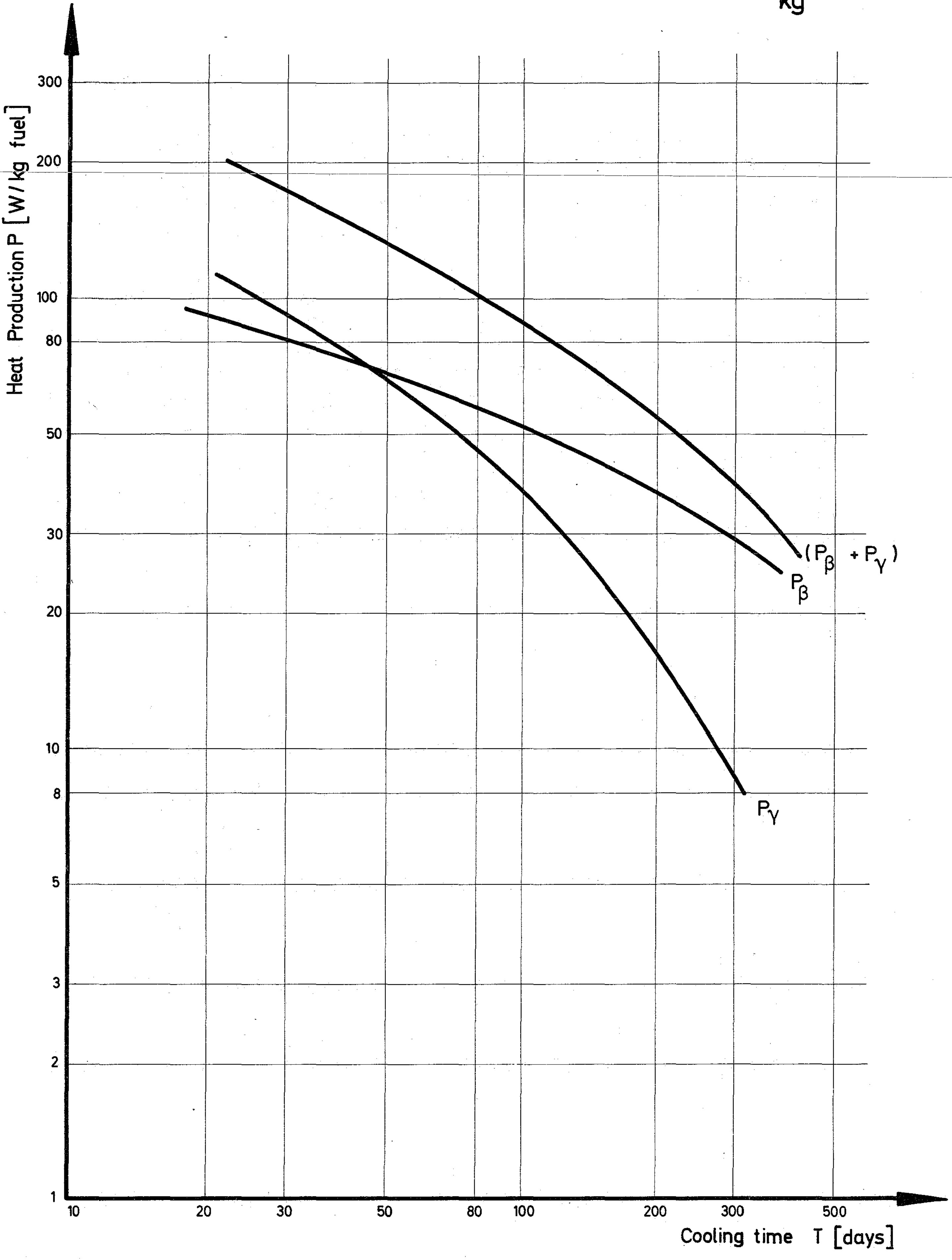

Fig. 2 Heat Production due to β - and γ - Decay for Reactor D1

in pile time 556 d burnup 56 $\frac{MWd}{kg}$

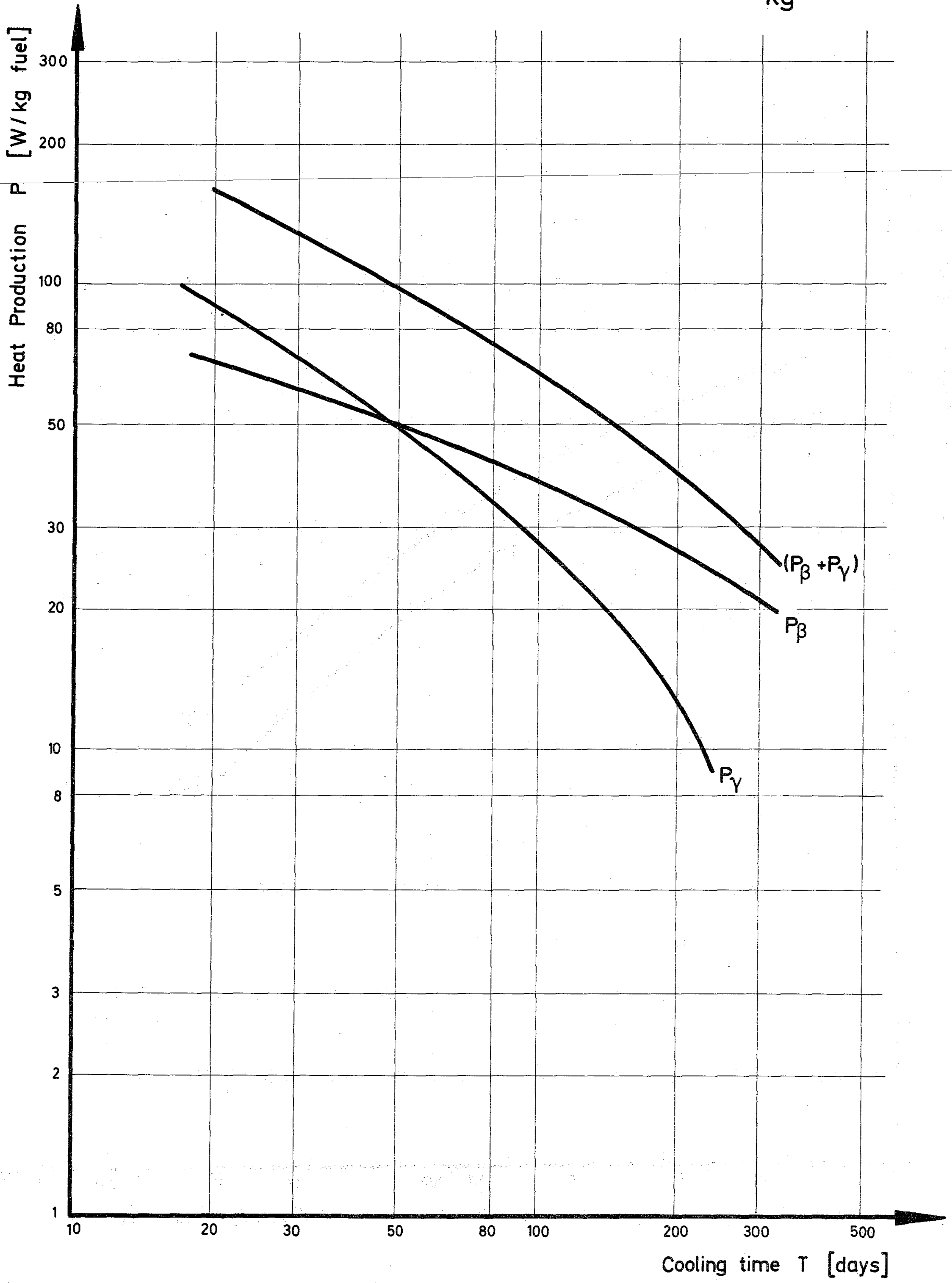

Fig. 3 Fissionproduct Activity of Na-1 and D-1 fuels

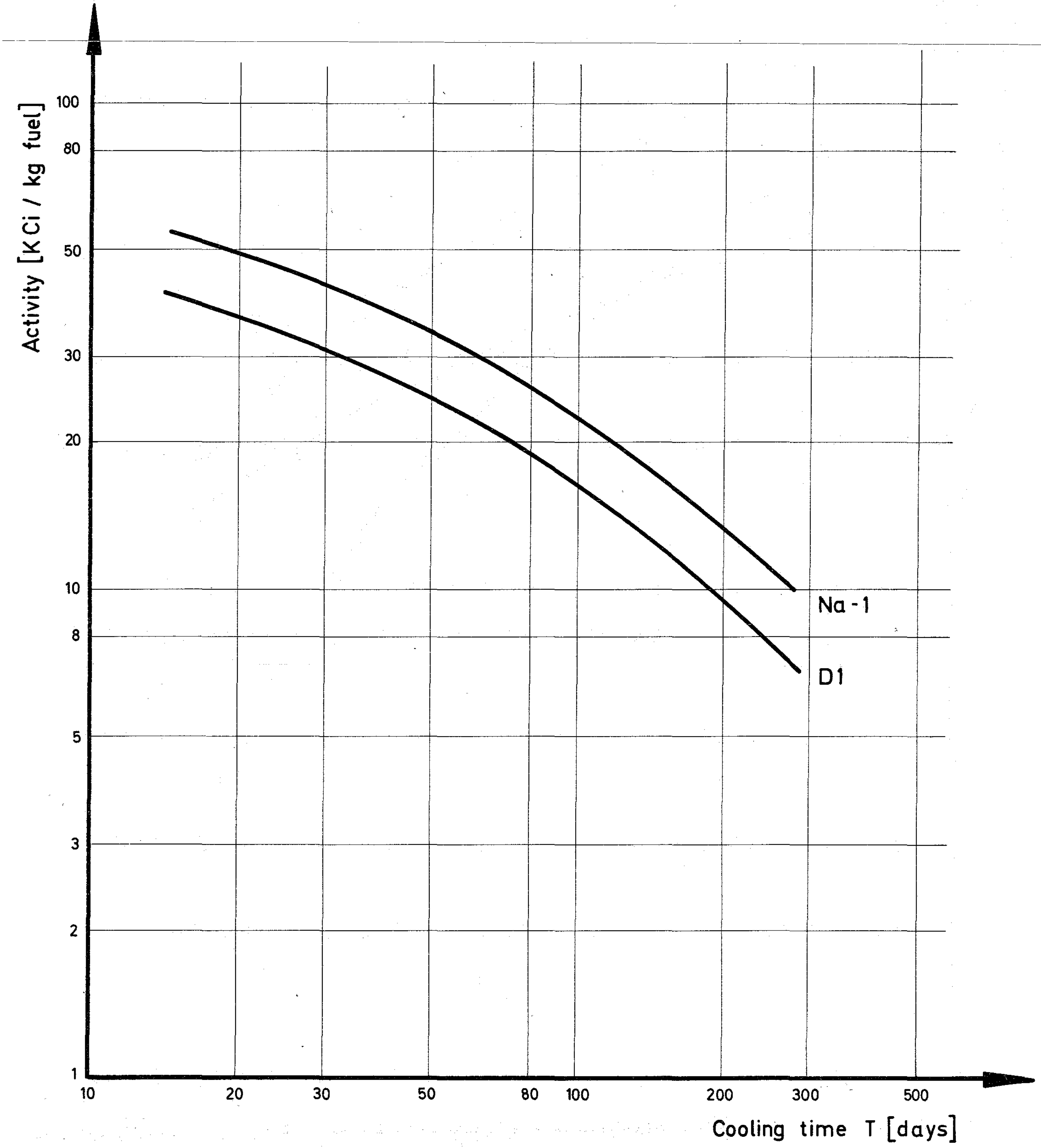

Fig. 4 Total Heat Production ($P_\beta + P_\gamma$) of Pu - Fuel for Various In - Pile Times

(1) King and Perkins (1958) Shure (1961)
(2) Stehn and Clancy (1958)