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THE DISTRIBUTION OF FISSION PRODUCT DECAY HEAT

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ABSTRACT

Data for the decay heat produced by each fission product element in a fast reactor are given for various times between 20 minutes and 4 months after shutdown of the chain reaction. These new data, obtained from the FISPIN code, are compared with the previous estimates and their relevance in PAHR studies examined. Following a core-meltdown it is estimated that at times less than a few hours 20% of the decay heating is by volatiles, while up to 30% may occur in a molten phase. After ten days the contribution of volatiles becomes negligible, but 20% of the heat input may still be in the steel debris.

Data on the total decay heating rate in both thermal and fast reactors are also reviewed. For post accident heat-removal studies, the Shure-Dudziak formulae as enshrined in the ANS standard continue to be useful. An empirical formula is presented which fits Phillips' FISPIN data. The simple $t^{-1/5}$ formula remains a good approximation in the period 1 hour to 1 year after shutdown.

(Paper presented at the Third Post Accident Heat Removal Conference, Argonne National Laboratory, Chicago, 3 November 1977)

INTRODUCTION

The total decay heat produced in a reactor after shutdown of the chain reaction is of fundamental importance in core debris control studies. Following a hypothetical accident in a fast reactor the heat sources may become distributed between the various phases present (e.g. vapour, liquid sodium, solid and liquid fuel debris, solid and liquid steel) depending on their volatility and chemical affinity, and so it is also necessary to know the breakdown of the decay heating rate by individual fission product elements to enable realistic assessments of the heat source in a particular phase or part of the reactor. In this paper we review the decay heat data currently in use for thermal and fast reactors and give the decay heat produced by each fission product element at times between 1000 seconds and $\approx 10^7$ seconds (113 days) according to the FISPIN code.

REVIEW OF DECAY HEAT DATA

The detailed dependence of the decay heat on time after shutdown depends on the irradiation history of the fuel elements, but differences only become significant when the elapsed time is of the same order as the mean time fuel has been in the reactor (i.e. typically a few hundred days for a fast reactor). In calculating the decay heat it is usual to consider the fission product inventory immediately before refuelling, or assume an infinite irradiation time and equilibrium concentration of all fission products. Data in this latter form may be readily applied to finite irradiation, provided flux-level effects are unimportant.

The decay heat consists of four terms: the heat from (i) fission products, (ii) the decay of actinides, (iii) irradiated reactor materials and (iv) fission (short times only).

It is assumed that all the energy in α , β and γ radiation is converted to heat within the core (or its debris). A substantial fraction of the decay energy is in the form of neutrinos, which are not absorbed in the reactor or elsewhere, and this energy is omitted in the calculations.

Some difference between the fission product contribution in fast and thermal reactors is likely as the fission product isotopes are formed with different probabilities, but large differences are neither found nor expected. According to Phillips' calculations (see below) fissions caused by the decaying neutron flux in a fast reactor produce a significant contribution to the heat in the first 100 seconds, but are insignificant at later times [1]. The contribution from (iii) and (iv) above depend on the particulars of the reactor system. The actinides do not make a significant contribution to the decay heat ($\lesssim 5\%$) in thermal reactor systems, but they do produce between 10 and 30% of the decay heat in a fast reactor. A further contribution to the heat in a sodium-cooled fast reactor is the activity of Na^{24} which has a half-life of 15 hours. As the decay heating rate for a fast reactor is unlikely to deviate by more than about 20% from that for a thermal reactor, data for the latter has been used in the past as much more work has been done for thermal systems [2].

Thermal Reactors

A great deal of work has been devoted to establishing fission product decay heat curves for thermal reactors (see Thompson & Beckerley (1973) [3] for a compact statement). The most used data sources are as follows:

- (i) The Way-Wigner (1948) [4] formula which is adequate for simple calculations:

$$P(t)/P_0 = \eta[t^{-0.2} - (T_0 + t)^{-0.2}] \quad (1)$$

where η is the Borst-Wheeler factor 0.0622, $P(t)$ is the rate of decay heating at time t seconds after shutdown, P_0 is the power on-stream and T_0 the period of reactor operation in seconds (partial refuelling may be considered easily by choosing appropriate values of T_0 and summing the contributions of the various fuel elements).

(ii) The Shure-Dudziak formulae [5], produced in 1961:

$$P(t)/P_0 = f(t) - f(t + T_0) \quad (2)$$

where $f(t) = (A/100)t^{-a}$; A and a are given in Table 1.

These formulae have been widely used, and taking $T_0 \rightarrow \infty$ (i.e. most conservative estimate) have been adopted as the ANS standard for times between 10^5 and 4×10^6 seconds. Shure (1972) [6] reviewed his 1961 recommendations in the light of more recent work and found them still in good agreement with authenticated data. Tobias [7] also found them extremely reliable when compared with later work.

(iii) The Untermeyer-Weills empirical formula (1962) [8].

(iv) Data (for periods up to 10^5 seconds) recommended by Teague (1965) [9] and used by Davies [2]. These agree with the ANS standard to better than 5% for times in excess of 300s.

A comparison of these decay heat curves is given in Fig.1 where infinite irradiation time is assumed. To show the effect of finite irradiation the Shure-Dudziak data are given for exposures of 10^7 seconds (117 days) and 10^8 seconds (3.2 years) in Fig.2. Decay heat data used in WASH-1250 for the period $10^5 - 3 \times 10^8$ seconds is also shown in Fig.2, where the assumed irradiation time is typical of LWR operation [10].

Fast Reactors

The data for fast reactors are much more limited. These include:

(i) Marr and Bunch (1971) [11], who calculated the decay heat for the FFTF using the RIBD code. They include a statistical sample of short-lived isotopes matching the decay heat to that found experimentally. Their decay heat data are for driver fuel and contain contributions from the β -radiation of ^{239}U and ^{239}Np involved in the transmutation of ^{238}U to ^{239}Pu . Alpha heating, from decay of the higher actinides, and contributions from breeding fuel are not included. They assess the errors on their curve as less than 20% for times in excess of 1000 seconds (less than 30% at shorter times).

(ii) Halliday (1972) [12], who reported two calculations for PFR: (a) one which included all heat sources, and in Halliday's report was increased by a 20% safety factor (omitted here), and (b) a summation using an AERE code which only included radiation from the most important fission products. This latter calculation was intended to determine the proportion of the heat from volatile elements etc. following a hypothetical core melt-down and, as the total decay heat found is on the low side, it was suggested that the results be used only in a comparative manner.

(iii) Phillips (1974) [1], who used an interim version of the FISPIN code, which includes over 350 isotopes, with input parameters applicable to the CFR design. Besides including the β -radiation from the actinides in the driver fuel Phillips also included contributions from the axial breeder, increasing the β -radiation from the actinides by 30% (about 6% of the reactor power comes from fissions in the axial breeder). He also investigated the effect of storage time of the fuel; heat is produced by the alpha decay of ^{242}Cm and ^{244}Cm . At times in excess of 3×10^5 s the alpha heating exceeds 10% of the fission product contribution for fuel stored for 4 years and is approximately proportional to the age of the fuel. At short times a contribution from fission was included, which is dependent on how the chain reaction is shut-down. An overall accuracy of 20% is claimed. The calculations are pessimistic in the sense that large fuel burn-ups are assumed (mean burn-up 7.5%). Phillips data may be approximated by power law segments of the form

$P(t)/P_0 = BT^{-b}$ where B and b are given in Table 2. This fit is normally better than 5% to the total rate of heating less that arising from decay of ^{24}Na . These formulae do not have the universality of the Shure-Dudziak data as they are not for infinite irradiation time.

(iv) Gluekler, contained in GE reports. An unreferenced decay heat curve is given in [13]. These data exceed those of other authors by more than 50% in places and it seems likely that a safety factor has been used. In a later report [14] another decay heat curve is given, referenced to a letter from Dickson to Riley (1973) on the Clinch River Breeder Reactor Plant. The heat produced by the β -decay of ^{239}Np is included and it is stated that this contribution raises the total decay heat as much as 70% above that from the fission products alone in the period 1 to 2 days after shut-down. (In Phillips' calculation ^{239}Np decay raises the total decay heat by 30% of the fission product contribution in this time period). No α -decay heating is included.

(v) Sporrer and Christenson (1975) [15], who used a modified version of England's CINDER code. It seems that only fission product decay heating was considered. In general the values found are greater than those given by other authors for the fission product contribution by 30-50%, although better agreement is found by comparing their data with the total decay heating found by other authors. Variations in irradiation history have the expected effects and they also find a flux level effect at long times after shut-down. The high rate of fission product heating according to this code ought to be verified.

Because build-up of the higher actinides in the fuel makes significant contributions to the decay heat in fast reactors it is not meaningful to consider the infinite irradiation limit (except for fission products alone) and so all curves are drawn for particular reactors and fuel cycles and must show the cut-off found in Fig.2 for thermal reactors and finite irradiation time. In Fig.3 the fission product contributions to the total decay heat according to Phillips, Gluekler [14], Halliday (a and b) and Sporrer and Christenson are compared with the thermal reactor cases of Shure-Dudziak with irradiation time of 10^8 seconds and with the Way-Wigner approximation assuming that one-seventh of the core is replaced every 47 days, as assumed by Halliday for PFR. In Fig.4 the total decay heating rates for the fast reactors considered by Halliday, Marr and Bunch, Phillips and Gluekler [14] are given together with Gluekler's [13] curve, and the Shure-Dudziak curve with irradiation time of 10^8 seconds for reference.

DISCUSSION - COMPARISON OF THERMAL AND FAST REACTOR DATA

The good agreement between the data recommended by Teague and the ANS standard (Shure-Dudziak data) for thermal reactors is to be emphasized. The Untermeyer-Weills curve (Fig.1) lies 40% above the ANS curve after 1 day and is almost certainly an over-estimate at these times. For periods less than 1 week the Way-Wigner formula is accurate to about 30%. The fission product contribution to the decay heat in a fast reactor may also be compared with the Shure-Dudziak data (Fig.3). All the fast reactor data except that of Sporrer and Christenson, lie below the Shure-Dudziak curve, the calculations of Gluekler (1975) and Halliday (b) being particularly low. Halliday (a) and Phillips show the closest agreement with thermal data (it is possible, however, that Halliday (a) is strongly influenced by thermal data). It is interesting to note that the Way-Wigner 'PFR' model fits most of the data to better than 20% for times in excess of 1000 seconds, although Fig.1 implies that it will give an over-estimate at very long times. Fig.4 shows that the effect of adding the other contributions to the decay heat in a fast reactor is to move the decay heat curve above the Shure-Dudziak thermal reactor curve for times less than 10^6 seconds. The amount of shift between Figs.3 and 4 varies for the different data sets. The data of Marr and Bunch lie below that of the others - it may be that as the FFTF is not designed as a breeder the contribution from β -decay of ^{239}U and ^{239}Np is much reduced. The data of Gluekler (1974) is consistently high. Uncertainties exist in the fast reactor case because of the importance of the β -radiation from ^{239}Np at intermediate times, which depends on the breeding ratio and whether heat from breeder elements is included. The alpha-heating discussed by Phillips adds further uncertainties.

INTEGRATED HEAT INPUT

In PAHR studies, data on the integrated decay heat are useful, as limits may be placed on the volume of materials melted and heat content of the debris etc. Although the rate of

decay heating falls with time the integrated heat per decade of time increases until the elapsed time is similar to the irradiation time. At longer times after shutdown the integrated heat $I(t, T_0)$ is a function of the irradiation time; $I(t, T_0)$ tending to $0.078 P_0 T_0^{4/5}$ according to the Way-Wigner approximation. The integrated decay heat data for various times after shutdown is shown in Table 3. The integrated decay heat for the Shure-Dudziak approximation is given for infinite irradiation time ($I(t, \infty)$); that for finite irradiation times follows simply: $I(t, T_0) = I(t, \infty) + I(T_0, \infty) - I(t + T_0, \infty)$. (Thompson and Beckerley provide a table of $I(t, T_0)$, based on the Shure-Dudziak approximation). Reasonable agreement is found between the different data sets except at long times where the assumed irradiation times dominate.

DECAY HEATING PER ELEMENT

The Calculation

Data from the FISPIN 3 code modelling of the fission product inventory of the CFR core (supplied by Broadley, Phillips and Burstall at NPC Risley) were used. The FISPIN code contains 343 fission product isotopes and 56 heavy element isotopes. The model assumes a fuel burn-up of 3.36% in 176 days where the fuel used was enriched by 25.5% AGR plutonium and stored for 4 years before burning. The estimated full thermal power of CFR on stream is 3223 MW. The predicted number of disintegrations per second of each isotope was multiplied by the energy per disintegration released as β and γ radiation (also taken from the FISPIN data set for consistency) to provide the decay heat per isotope per second. Hand summation of the heat per isotope led to the heat per atomic species per second, and these are given in Table 4 as fractions of the full thermal output power. Data are given for each decade of time between 10^3 and 10^7 seconds after shutdown. The actinides producing α , β and γ radiation, are incorporated in the FISPIN code but have not been treated individually here as they are likely to remain with the fuel debris. The contributions have not been split into β and γ components, but it should be noted that the heating effect of the β radiation is more localized than that produced by the γ s; β radiation accounts for approximately two-fifths of the total heating.

Application to PAHR Studies

In a severe reactor accident the fuel and cladding will melt allowing migration of the fission products. The data given in Table 4 may be combined with chemical and physical data on the fission products to predict the distribution of the heat source following the accident. Such estimates have been made by Booth [16] and Halliday [12]; work reported by Gluekler [17], the experiments of Fischer et al. [18] and the predictions of Gabelnick & Chasanov [19] are also relevant. It is usual to predict the chemical form of each fission product element at melt-down temperatures (besides the above references, a recent investigation by P E Potter (private communication) has been used below) and then investigate their volatility, and this procedure is followed below.

There is general agreement that (i) the noble gases (Kr and Xe) will escape to the cover gas, (ii) the halogens (Br and I) will form sodium halides and thus be removed from the debris, (iii) that Rb, Tc, Ru, Rh, Pd, Ag, Cd, In, Sn, Sb, Te and Cs will remain in elemental form, (iv) that Sr, Y, Zr, Ba, La, Ce, Pr, Nd, Pm and Sm will be present as oxides.

This leaves Nb and Mo unplaced. Mo and Nb act as buffers for the oxygen potential of the system. Fischer et al. found that in (a) the molten iron-molten UO_2 system and in (b) the molten iron-molten UO_2 /basalt system, Mo is present as the element in the molten iron phase, while Nb was distributed as both oxide and element in system (a) but occurred only as the oxide in the molten UO_2 /basalt mixture in system (b).

A preliminary division of the decay heat may be made according to points (i) to (iv) above and volatility effects considered separately. The decay heats summed in this way are given in Table 5, and shown in Fig.5 as percentages of the total decay heat. At short times (1000 seconds) nearly 20% of the decay heat is produced by the noble gases and halogens, and 30% by fission products in elemental form (including Mo), after 1 day the contribution of the latter group has dropped to 13% of the total, but it returns to around 20% at longer times. After 13 days the noble gases and halides provide less than 5% of the total decay heat. At times in excess of 1 day the fission product oxides and actinides (also oxides) produce between 65% and 75% of the total decay heat.

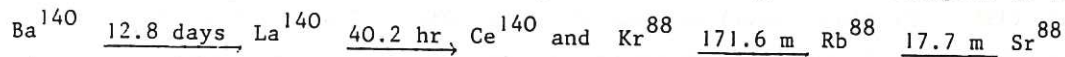
In general it is expected that the oxides and actinides will remain with the fuel debris and the fission products which remain in elemental form migrate to the steel debris or form a metallic phase included in the oxide phase.

The experiments of Fischer et al. provide confirmation of these ideas. Some fission products may be too volatile to remain in the molten debris, but consideration of this effect is hampered as some of the boiling points, particularly of the oxides, are not known to sufficient accuracy: for instance the boiling point of BaO is given variously as 2000°C and 2750°C. The elements listed in (iii) may be split for convenience into groups based on their boiling point: (iiiA) Boiling point between 600°C and 900°C Cs, Rb, Cd, (iiiB) Boiling point between 900°C and 1400°C Te, Sb, (iiiC) Boiling point between 1400°C and 2300°C In, Ag, Sn, (iiiD) Boiling point in excess of 2800°C Tc, Ru, Rh, Pd (and Mo, and Nb if in elemental form).

This classification includes all the relevant fission products. The decay heat from groups (iiiA) - (iiiD) is given in Table 6. The higher boiling point groups will probably remain with the steel, the lower boiling point group(s) migrating to the sodium or vapour phases.

The stable oxides (listed (iv) above) all have high boiling points; BaO is probably the most volatile, if the lower boiling point of 2000°C is used, while La₂O₃ and ZrO₂ have boiling points in excess of 4000°C, the other oxides boiling at intermediate temperatures. The decay heat data for the oxides is split into three groups on this basis (see Table 6).

The above scheme may lead to incorrect conclusions because a less volatile daughter element may be produced by a volatile fission product. Two significant examples of this are:



both La¹⁴⁰ and Rb⁸⁸ are the most important fission products of their respective elements as far as decay heating is concerned, and so after 18 minutes for Rb and 40 hours for La most of their decay heat should be added to the phase in which the parent isotope is found unless further migration occurs.

The general conclusions from the data in Table 6 are that (a) volatility of group (iii) elements is more important than that of the group (iv) oxides. (b) volatility may reduce the heating rate in the steel by as much as 50% at short times, but by less than 10% at times in excess of 10 days. (c) if the temperature of the fuel debris can be maintained below about 2600°C (e.g. by mixing with a sacrificial material) volatility of the oxide fission products is small except possibly for BaO and its effect on La₂O₃.

Many aspects of the distribution of fission product heating have not been covered in the above discussion such as: (i) the equilibrium distribution of fission products between sodium and steel. (ii) possible plate out of oxides on steel surfaces suggested by Halliday [12]. (iii) the behaviour of carbide fuel. (iv) the effect of sacrificial material, except that basalt and by implication concrete do not affect the distribution between an oxide and a metal phase. Should UO₂/PuO₂ fuel melt the sacrificial material, and then, after cooling, solidify out, the distribution of fission products between the solid and molten phases is unknown.

The data given in Table 4 may be used with appropriate chemical data to investigate these points.

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Applicable Time Interval (sec)	A	a
$10^{-1} < t < 10^1$	6.02	0.0639
$10^1 < t < 1.5 \times 10^2$	7.66	0.1807
$1.5 \times 10^2 < t < 4 \times 10^6$	13.01	0.2834
$4 \times 10^6 < t < 2 \times 10^8$	26.59	0.3350

TABLE 1. Power-law fit to Shure-Dudziak data (Infinite irradiation)

Applicable Time Interval (sec)	B	b
$1 < t < 3 \times 10^5$	0.137	0.285
$3 \times 10^5 < t < 2 \times 10^7$	3.3	0.54
$2 \times 10^7 < t < 10^8$	3.8×10^6	1.37

TABLE 2. Power-law fit to Phillips' data (CFR model)

TIME (SEC)	W - W(4)	THERMAL U - W(8)	S - D(5)	S - C(15)	FAST P(1)	M - B(11)
10^3	19	27	24	30	27	21
10^4	120	170	130	180	140	110
10^5	780	950	700	990	780	560
10^6	4900	4900	3600	5100	3500	2500
10^7	3.1×10^4	2.1×10^4	1.8×10^4	1.7×10^4	1.2×10^4	8600
10^8	2.0×10^5	7.3×10^4	8.3×10^4	3.3×10^4	2.5×10^4	1.8×10^4

TABLE 3: Integrated decay heat. 1 unit is the energy delivered by the reactor in 1 second when fully operating. Note that thermal data are for infinite irradiation times but fast data are for specific reactor cycles. The effect of varying reactor operating time is shown in Table 4-5 of Thompson and Beckerley [3].

TABLE 4

TIME/ Element	10^3 s	10^4 s	1 DAY	13 DAYS	113 DAYS
Br	0.01	0.	0.	0.	0.
Kr	2.69	1.51	0.01	0.	0.
Rb	4.22	1.43	0.01	0.	0.
Sr	6.20	3.44	0.83	0.39	0.10
Y	12.44	5.93	1.45	0.60	0.18
Zr	5.17	4.82	2.95	1.42	0.49
Nb	7.18	5.82	3.13	1.36	0.81
Mo	5.04	1.77	1.41	0.07	0.
Tc	13.07	0.38	0.30	0.01	0.
Ru	4.60	3.48	1.61	1.25	0.22
Rh	2.82	1.71	1.50	1.07	0.83
Pd	0.65	0.53	0.11	0.	0.
Ag	0.44	0.38	0.21	0.02	0.
Cd	0.12	0.07	0.02	0.	0.
In	0.11	0.04	0.	0.	0.
Sn	0.71	0.25	0.08	0.04	0.01
Sb	5.63	0.92	0.20	0.03	0.
Te	9.31	2.53	1.17	0.11	0.01
I	27.42	18.75	8.74	0.80	0.
Xe ¹	5.48	2.75	1.22	0.11	0.
Cs	11.90	0.57	0.04	0.02	0.01
Ba	7.60	2.09	1.27	0.67	0.02
La	19.44	11.66	7.35	4.17	0.02
Ce	2.80	2.32	1.70	0.51	0.11
Pr	6.62	2.36	1.57	1.18	0.61
Nd	1.28	0.68	0.43	0.20	0.
Pm	0.61	0.57	0.38	0.01	0.01
Sm	0.	0.	0.	0.	0.
TOTAL FISSION PRODUCTS	163.5	76.8	37.7	14.03	3.43
ACTINIDES ²	25.1	15.5	12.4	3.09	1.87
TOTAL DECAY HEAT	188.6	92.3	50.1	17.1	5.30

Decay heat per element at various times after shutdown, in units of 10^{-4} x full thermal power of reactor. FOOTNOTES: 1 Revised figures (original FISPIN figures for Xe are 6.35, 3.62, 1.22, 0.11 and 0). 2 Does not include breeder fuel. Phillips increases the β component by 30% to allow for axial breeder.

TABLE 5

Decay heat by groups of elements at various times after shutdown expressed (a) units of 10^{-4} x full thermal power of reactor and (b) percentages of total decay heat.

	10^3 seconds		10^4 seconds		1 DAY		13 DAYS		113 DAYS	
	(a)	(b)%		%		%		%		%
Kr + Xe (i)	8.17	4.33	4.26	4.61	1.23	2.46	0.11	0.6	0.	0.
Br + I (ii)	27.43	14.54	18.75	20.32	8.74	17.45	0.80	4.7	0.	0.
Elements (iii)	53.58	28.41	12.29	13.31	5.25	10.49	2.55	14.9	1.08	20.4
Mo	5.04	2.67	1.77	1.92	1.41	2.82	0.07	0.4	0.	0.
Nb	7.18	3.81	5.82	6.30	3.13	6.25	1.36	7.9	0.81	15.3
Oxides (iv)	62.1	32.93	33.9	36.72	17.9	35.76	9.14	53.4	1.54	29.1
Actinides	25.1	13.31	15.54	16.83	12.4	24.77	3.09	18.0	1.87	35.3
TOTAL	188.6	100	92.3	100	50.1	100	17.1	100	5.30	100

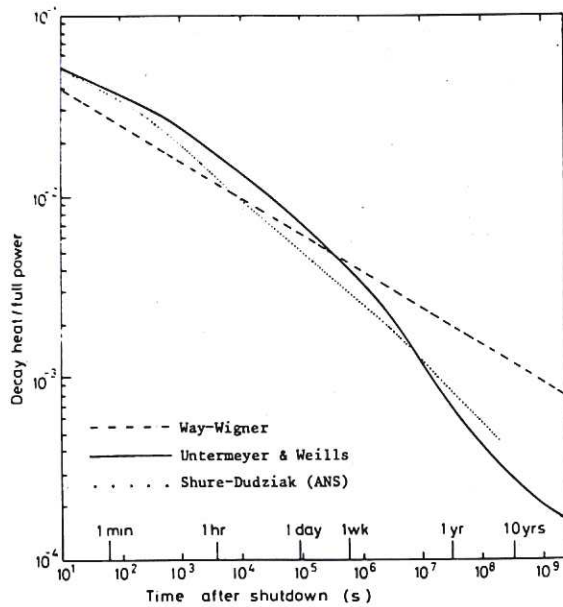


FIG. 1. Decay heat for thermal reactor. Infinite irradiation.

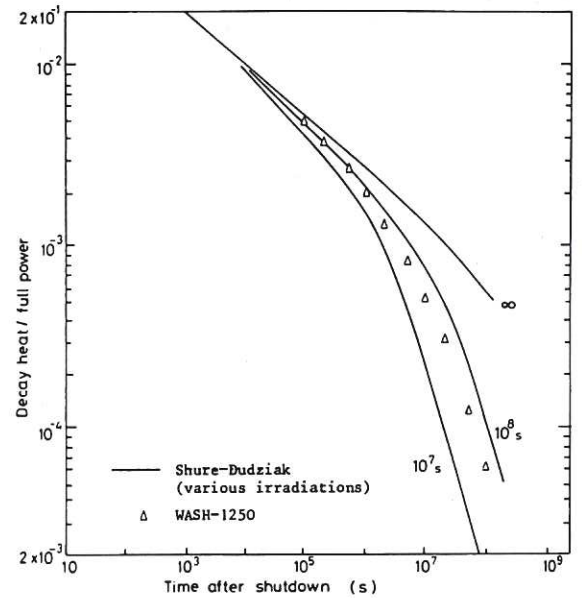


FIG. 2. Decay heat for thermal reactor. Finite irradiation.

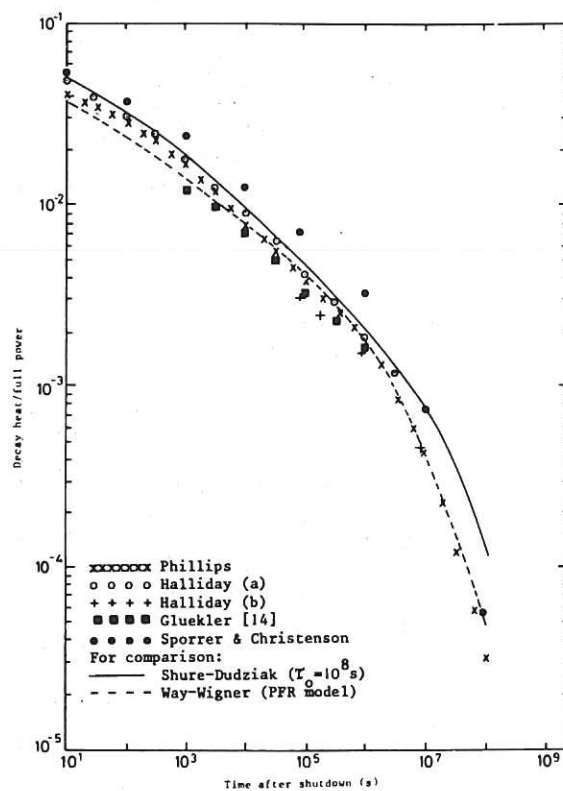


Fig. 3. Decay heat for fast reactor. Fission product heat only.

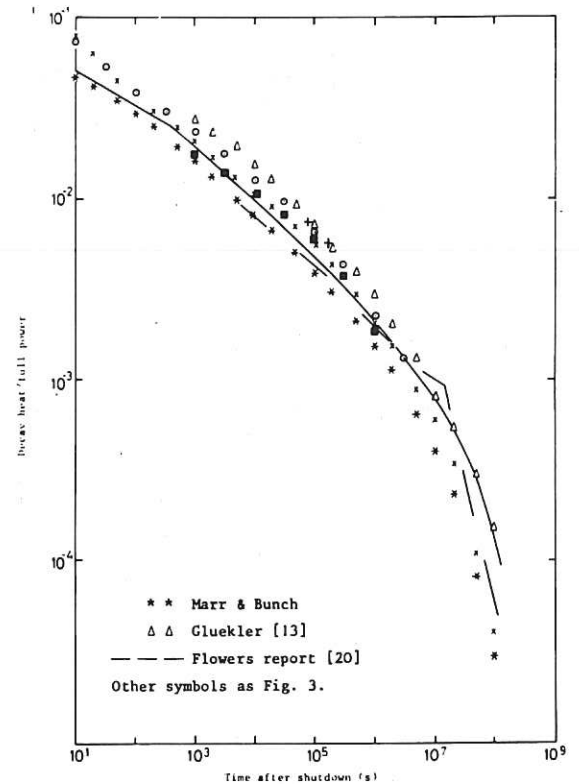


FIG. 4. Decay heat for fast reactor. Total decay heat.

TABLE 6

		10 ³ seconds	10 ⁴ seconds	1 day	13 days	113 days
Elements:	(iii A)	16.24	2.07	0.07	0.02	0.01
	(iii B)	14.94	3.45	1.37	0.14	0.01
A + B		31.18	5.52	1.44	0.16	0.02
	(iii C)	1.26	0.67	0.29	0.06	0.01
A + B + C		32.44	6.19	1.73	0.22	0.03
	(iii D)	21.14	6.10	3.52	2.33	1.05
TOTAL		53.58	12.29	5.25	2.55	1.08
	Nb	7.18	5.82	3.13	1.36	0.81
	Mo	5.04	1.77	1.41	0.07	0.0
Oxides:	Ba	7.60	2.09	1.27	0.67	0.02
Intermediate boiling points		29.9	15.33	6.43	2.88	1.01
Zr & La ¹		24.61	16.48	10.20	5.59	0.51
TOTAL		62.1	33.9	17.9	9.14	1.54

TABLE 6: Breakdown of decay heat according to relative volatility. If Nb and Mo are in elemental form they should be added to (iii D), if in oxide form to the intermediate boiling points group. FOOTNOTE¹: See note in text on location of La₂O₃.

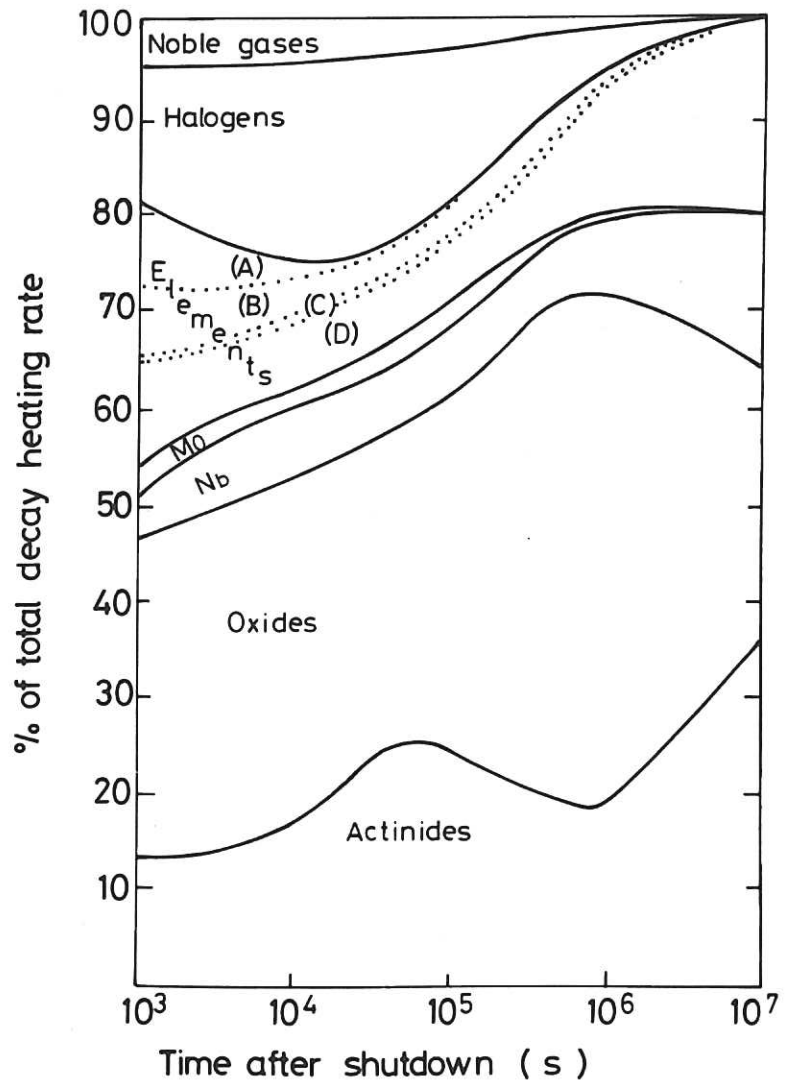


FIG.5: Cumulative contributions to the decay heating rate. Following a meltdown the actinides and the 'oxide' group are expected to remain with the fuel debris while the 'elements' group will probably migrate to the steel debris. The elements are subdivided on the basis of volatility as described in the text. Volatile elements are shown at the top of the figure.

