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Research report
NFL-12
1979

UNCERTAINTY OF DECAY HEAT CALCULATIONS ORIGINATING FROM ERRORS IN THE NUCLEAR DATA AND THE YIELDS OF INDIVIDUAL FISSION PRODUCTS

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SE7900292

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by

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Abstract

The calculation of the abundance pattern of the fission products with due account taken of feeding from the fission of ^{235}U, ^{238}U, and ^{239}Pu, from the decay of parent nuclei, from neutron capture, and from delayed-neutron emission is described. By means of the abundances and the average beta and gamma energies the decay heat in nuclear fuel is evaluated along with its error derived from the uncertainties of fission yields and nuclear properties of the individual fission products. (author)

1. INTRODUCTION

In recent years a new technique has almost revolutionized the study of short-lived fission products. This is the ISOL-technique, i.e. attaching an isotope separator on-line with an accelerator or a reactor. A powerful example of this technique is the OSIRIS facility at Studsvik[1,2] where a great many neutron-rich activities have been studied, more than 50 of them for the first time. Thus, our knowledge about the decay characteristics of short-lived fission products has grown considerably, and it seems worth while to make a new evaluation of the abundance pattern of fission products in nuclear fuel with emphasis on the short-lived ones. To this end a code "INVENT" has been developed taking into account transfer between mass chains not only by neutron capture but also by the emission of delayed neutrons which is an important mode of decay far from stability. Another feature is that track is kept of the uncertainties of the various decay and yield data so that the accuracy of the final results can be stated.

Input data in the present version of the programme are fission yields from ^{235}U, ^{238}U, and ^{239}Pu (for reactors using fuel containing other fissionable materials, the code can be modified), half-lives, neutron capture cross sections, internal transition rates in case of isomers, and average beta and gamma energies. One isomeric state in addition to the ground state may be taken into account for each nuclide (occasionally more than one isomer of reasonably long half-life may be found; in such a case the most important two states are chosen for the nuclidic library).

The knowledge of the distribution of fission products in nuclear fuel as a function of the fuel composition and the irradiation conditions is of great importance with applications in various fields of nuclear technology. Examples are the reprocessing of fuel and the treatment of radioactive waste. In these cases it is sufficient to know the distribution of long-lived products. For many applications the short-lived ones are also very important, however. An example of this is the evaluation of the power developed in the fuel at short times after stopping a nuclear reactor. The reason for this is that the highest independent fission yields are found for very neutron-rich nuclides far from stability. At saturation the decay rate of all the isobars closer to stability in a given mass chain will be roughly equal to the decay rate of the short-lived isobars of maximum independent yield. As those isobars normally possess the highest Q_β-values, their contribution to the decay heat will be larger than that of the more long-lived species. Naturally, this situation will rapidly change with time as the short-lived nuclides die out, but in connection with estimates of the heating of the fuel in case of a reactor accident (loss-of-coolant accident) one is concerned about the power developed at short times after the accident and thus by the decay of the short-lived fission products.

Other examples are the evaluation of the energy spectra of beta particles, gamma rays, and delayed neutrons at equilibrium and as a function of the cooling time

after shutting down a reactor. Still another application, this time in fundamental research rather than in nuclear technology, is the evaluation of the anti-neutrino spectrum around a reactor, especially its high-energy part[3]. The interest here stems from the fact that the nuclear reactor is an excellent source of anti-neutrinos used in various experimental work (cf.,for instance, ref.[4]).

A number of codes have been developed for calculating the decay heat by the summation method. These have been covered in a recent review by Schenter, Schmittroth, and England[5]. In some cases the uncertainty of the final results has been derived by an evaluation of the sensitivity of the calculation to variations of certain of the parameters involved[6-10]. The present approach is more complete following the propagation of the errors of the physical quantities appearing in the calculation - decay constants (including branching ratios), capture cross sections, average beta energies, average gamma energies, delayed neutron branching ratios, and fission yields - giving the uncertainty of the final result with an analysis of the composition of this uncertainty in terms of contributions from the various types of quantities listed above. As an option, the contribution of each individual fission product to the total decay heat, together with an analysis of its error, can be obtained. The reason for this rather extensive error analysis is to provide a method for finding out which nuclear data need be improved and for demonstrating the effect of such improvements on the decay heat evaluation. This is of great importance for the planning of experiments on short-lived fission procucts at this laboratory, and it should also be of interest for other groups involved in similar research projects.

2. MATHEMATICAL TREATMENT

Only the main lines of the mathematical treatment are given in the present note. A full account is found in a laboratory report[11], which is available and to which the reader is referred for details.

2.1 The actinides

The first step in the calculation is to evaluate the time dependence of the abundance of the actinides. For ^{235}U and ^{238}U the treatment is straight-forward - these materials are simply used up by neutron capture and by fission. The third fissionable material taken into account, ^{239}Pu, is formed from ^{238}U by the following chain (and may also be present in the original fuel):

$$^{238}U + n \rightarrow {}^{239}U \xrightarrow{\beta^-} {}^{239}Np \xrightarrow{\beta^-} {}^{239}Pu.$$

This case is also treated in a straight-forward manner.

The nuclide ^{239}Pu can also be formed from ^{235}U via the secondary route:

$$^{235}U + n \rightarrow {}^{236}U + n \rightarrow {}^{237}U \xrightarrow{\beta^-} {}^{237}Np + n \rightarrow {}^{238}Np \xrightarrow{\beta^-}$$
$$^{238}Pu + n \rightarrow {}^{239}Pu.$$

This route is usually of much less importance, and it is neglected here. Furthermore, neutron capture and beta decays starting from ^{239}Pu will yield a number of actinides up to ^{244}Cm, among them the fissionable ^{241}Pu and ^{242}Am. Since the main aim of the present work has been to study the behaviour of the fission products these additional actinides have not been included in the computer code. If necessary, the programme can be extended to include the more important ones of these nuclides.

The errors of all the abundances of the actinides appearing in the treatment are evaluated by quadratic summation of the errors of all the physical quantities used in the calculation.

2.2 The fission products. First approximation

The general differential equation governing the variation of the amount N(Z,A,X) of the nuclide (Z,A,X) of atomic number Z and mass number A and where X stands for L (= ground state) or H (isomeric state) may be written:

$$\frac{dN(Z,A,X)}{dt} = -\left[\lambda(Z,A,X,H) + n \times \sigma_c(Z,A,X,H) + \lambda(Z,A,X,L) + \right.$$

$$\left. + n\times\sigma_c(Z,A,X,L) + \delta(X-H) \times \lambda(Z,A,I)\right] \times N(Z,A,X) + n \times$$

$$\times N(92,235) \times \sigma_f(92,235) \times Y_5(Z,A,X) + n \times N(92,238) \times$$

$$\times \sigma_f(92,238) \times Y_8(Z,A,X) + n \times N(94,239) \times \sigma_f(94,239) \times$$

$$\times Y_9(Z,A,X) + \lambda(Z-1,A,L,X)\left[1-P_n(Z-1,A,L,H) - P_n(Z-1,A,L,L)\right] \times$$

$$\times N(Z-1,A,L) + \lambda(Z-1,A,H,X) \times \left[1-P_n(Z-1,A,H,H) - \right.$$

$$\left. - P_n(Z-1,A,H,L)\right] \times N(Z-1,A,H) + \delta(X-L) \times \lambda(Z,A,I) \times N(Z,A,H) +$$

$$+ \left[\lambda(Z-1,A+1,H,H) + \lambda(Z-1,A+1,H,L)\right] \times P_n(Z-1,A+1,H,X) \times$$

$$\times N(Z-1,A+1,H) + \left[\lambda(Z-1,A+1,L,H) + \lambda(Z-1,A+1,L,L)\right] \times$$

$$\times P_n(Z-1,A+1,L,X) \times N(Z-1,A+1,L) + n \times \sigma_c(Z,A-1,H,X) \times$$

$$\times N(Z,A-1,H) + n \times \sigma_c(Z,A-1,L,X) \times N(Z,A-1,L), \qquad (1)$$

with the notations

n = neutron flux,

$\sigma_f(Z,A)$ = fission cross section for the nuclide (Z,A),

$Y_j(Z,A,X)$ = independent fission yield of the nuclide (Z,A,X) with $j = 5, 8, 9$ for ^{235}U, ^{238}U, and ^{239}Pu, respectively,

$\lambda(Z,A,X,Y)$ = decay constant of the nuclide (Z,A,X) leading to the state Y of the daughter,

$\lambda(Z,A,I)$ = decay constant for internal transition in (Z,A,H),

$P_n(Z,A,X,Y)$ = neutron branching ratio of the precursor (Z,A,X) leading to the state Y of the final nucleus,

$\sigma_c(Z,A,X,Y)$ = neutron capture cross section of (Z,A,X) leading to the state Y of the product,

$\delta(X-Y) = 1$ for $X = Y$ and $= 0$ for $X \neq Y$.

X and Y stand for L (ground state) or H (isomeric state).

For the determination of the fission rates and the neutron capture rates a somewhat modified Westcott formalism is used taking into account the neutron spectrum at the reactor temperature, the Westcott g-factor, the

thermal neutron capture cross section, the resonance integral, the epithermal flux, and a spectrum parameter.

The system of equations (1) may be solved directly, but the solution would be very cumbersome because the amount of any product will depend on the nuclear data (i.e. decay constant, neutron capture cross section, fission yield, etc.) of all other fission products and of the actinides treated in Section 2.1. An approximate but quite accurate solution can be found, however, by neglecting the feed via delayed-neutron emission and neutron capture. This means a very considerable simplification because the abundance of a fission product (Z,A) will, in this approximation, be a function only of the decay data of isobars of the same mass number with atomic number $\pm$ Z and of the actinides ^{235}U, ^{238}U, ^{239}U, ^{239}Np, and ^{239}Pu. The exact solution can then be written down as soon as a lower atomic number $Z_o(A)$ is defined as the first isobar of mass number A to be taken into account, i.e. whose fission yield exceeds some small value. Isobars still further out from stability are neglected.

2.3 The fission products. Second approximation

A further step towards the solution of the equation system (1) is obtained in the following way. The approximate solution from the preceding section is applied to one mass number below and one above the mass to be considered. The resulting abundances are then entered into the terms corresponding to feeding by neutron capture and delayed emission in Eqs. (1), and these equations are solved exactly to give the second approximation for N(Z,A,X). This approximation is expected to be very accurate because the first approximation is already quite good and, furthermore, it is only used in small correction terms. The term corresponding to delayed-neutron emission is always small. Close to stability the P_n-values are small or zero and further away the fission yields drop so that N(Z-1,A+1,X) will be much smaller than N(Z,A,X). The neutron capture term may occasionally

be large (cases with large resonance capture). However, large capture cross section in two adjacent nuclides, one and two mass numbers below the one under study and belonging to the same capture chain, are needed to produce an appreciable error, and this is not very probable. Among the nuclides which might be somewhat affected are ^{153}Sm, ^{157}, ^{158}Gd, and $^{163-166}Dy$ whereas cases such as ^{133}Cs and ^{135}Xe with large capture cross sections do not give rise to errors because the capture cross sections of ^{134}Cs and ^{136}Xe are not large enough.

Note that already the first approximation is the exact solution for zero neutron capture and zero delayed-neutron branching ratio and very close to the exact solution for small values of these quantities. Only for cases with large capture cross sections or large delayed-neutron branching ratios the difference between the first and the second approximations exceeds the uncertainty of the calculated results. Thus, the conclusion may be drawn that the procedure adopted will yield accurate N(Z,A,X)-values.

The technique for the evaluation of the time variation of the amounts of the various fission product in nuclear fuel exposed to a constant neutron flux is now established. The fact that the independent fission yields are used in the formulae makes them unsuitable for evaluating the variances of the abundances, however, the reason being that the errors of the independent yields are often very large. Instead, a different approach is used for the error analysis. The abundance of a given nuclide (Z,A,X) can be approximately obtained by adding, to the amount directly formed, contributions from isomeric transition, from beta decay, from delayed-neutron emission, and from neutron capture involving the nuclides (Z,A,H), (Z-1,A,L and H), (Z-1, A+1,L and H), and (Z,A-1, L and H), respectively. The abundances of the latter four types of nuclides are calculated using their cumulative yields but disregarding feeding from other fission products. The errors are evaluated taking into account the uncertainties of the independent yields (for ^{235}U, ^{238}U, and ^{239}Pu) of the fission product under consideration, those of the cumulative yields of the contributing

nuclides mentioned above, and furthermore the uncertainties of the half-lives, the delayed-neutron branching ratios, and the neutron capture cross sections of the nuclides involved. Although this error analysis is simplified it still contains a large number of contributions, or 46 for isomeric states and 47 for ground states (of which some may be zero).

The method above is used to calculate the abundances and their variances, at the end of an irradiation period with constant neutron flux. These values are then used as input for a calculation for the next period, and the procedure is repeated until the end of the irradiations is reached. The same scheme is, in fact, used also during the cooling time with the flux then put equal to zero. In this way the fuel composition at any time during the irradiation - cooling cycle is computed and can be tabulated.

In the special application of decay heat this quantity is obtained as a sum of contributions, one for each fission product.
Thus,

$$\text{decay heat} = \sum_i \lambda_i N_i(t) \; (\bar{\beta}_i + \bar{\gamma}_i), \qquad (2)$$

where $\bar{\beta}_i$ and $\bar{\gamma}_i$ are the average beta effect and gamma effect per disintegration of the product i. The effect of the actinides, as far as they are taken into account by the programme (cf. Section 2.1), may also be included.

Other quantities may be computed, such as the intensity and energy spectrum of the delayed neutrons, the composite beta and gamma spectra, etc. If such quantities are required, the abundances, and their variances, are stored on magnetic tape to be combined with the appropriate spectra for individual fission products in a subsequent calculation.

2.4 Constant power approximation

As an option a constant power can be used instead of a constant flux for the irradiation periods. This power is taken to be proportional to the fission rate. It is obtained from the integral

$$\text{Power} = \frac{n}{T} \int_0^T \{\sigma_f(92,235)\cdot N(92,235)\cdot E_5 + \sigma_f(92,238)\cdot N(92,238)\cdot E_8 + \sigma_f(94,239)\cdot N(94,239)\cdot E_9\}\, dt, \qquad (3)$$

where E_5, E_8, and E_9 are the total energies developed per fission of ^{235}U, ^{238}U, and ^{239}Pu, respectively.

Obviously, the amounts of fissionable nuclei also depend on the neutron flux $\underline{n}$. The value of $\underline{n}$ giving the required fission rate, or power, as an average over the irradiation period of length $\underline{T}$ can be deduced by an iterative method, however, using as the first approximation the value of $\underline{n}$ obtained with the initial amounts of ^{235}U, ^{238}U, and ^{239}Pu inserted into Eq. (3). The second approximation is obtained by evaluating the integral of Eq. (3) with the first approximation of the flux in the formulae for N(Z,A). In this way the code proceeds until the relative difference between the successive fluxes is smaller than 1 %. The corresponding flux is then used for the calculation of the abundances of the fission products.

3. DATA LIBRARY

3.1 Half-lives

The mass range used for the fission products is confined to 71 - 167 as fission products outside of this range have negligible yields. Only known fission products have been entered into the data library "FPLIB". Because of powerful experimental techniques the number of known nuclides is now high, and the data library contains 487 fission products and 5 actinides. In fact, the sum of the independent yields of this set of 487 fission products amounts to 98.8 % of the total fission yield for ^{235}U and to 99.2 % for ^{239}Pu, whereas the figure for fast fission of ^{238}U is somewhat lower, or 95.4 %. The latter fuel component is much less important than the others, however, and one can safely state that the effect of fission products, still unknown, on integral properties must be almost negligible.

The fission products included in the library are indicated in Table 1.

References[12-35] in the reference list have been used for the half-lives.

3.2 Neutron capture gross-section

Sources for thermal neutron capture cross sections (2200 m/s) and resonance integrals are the references[36-39]. Only measured cross sections are included in the library. In cases where no error limits are given in the references those have been estimated from the spread of measured values (unweighted). If only one measured value is published, the error limit has been arbitrarily put equal to 30-50 % of the cross section.

3.3 Delayed-neutron branching ratios

All branching ratios are taken from ref.[19]. Certain nuclides are known to be delayed-neutron precursors, but no branching ratio has been determined so far. To this group belong: $^{79-82}Ga$, ^{83}Ge, and $^{129,130}In$. They are neglected until experimental results become available.

3.4 Fission yields

The main source for the fission yields is the compilation by Rider and Meek[40]. In this compilation the fine structure in the isobaric yield curves has been taken into account. More extensive experimental investigations are needed, however, for an improved evaluation of this effect.

Wherever available, experimental fission yields have been included in the library[41-46].

Normally the independent yields are used in the library. An exception is the first member of each chain, for which the cumulative yield has been chosen instead.

The sum of all the yields for the fission products in the mass range 71 - 167 is 200.3 % for thermal-neutron induced fission of ^{235}U. The corresponding figures for fast fission of ^{238}U and thermal fission of ^{239}Pu are 200.8 % and 199.3 %, respectively. The reason for the deviation from 200 % is due to the fact that experimental fission yields are mixed with calculated ones. A correction is built into the programme which adjusts the yields by multiplying them by the ratio 200/(sum of yields).

3.5 Average beta and gamma energies

The determination of average beta energies is the goal of an extensive experimental programme at this laboratory[47]. The aim is to get accurate values for all the products obtained at OSIRIS which means that experimental determinations are possible for nuclides whose independent yields amount to more than 80 % of the total fission yield.

Average beta and gamma energies can also be calculated from beta feed functions (the beta-branching as a function of the excitation energy of the daughter nucleus). Such beta feed functions can be derived from published decay schemes. Others are obtained from an experimental study of the beta strength properties of neutron-rich nuclides[48]. For cases where no experimental basis for a determination of

the average energies exists, the beta feed functions have been estimated from an extrapolation of the beta strength results.

The calculated average beta and gamma energies have been combined with directly determined average beta energies into a set of data for the present nuclidic library[47].

4. EXAMPLES FROM THE USE OF THE INVENTORY CODE FOR DECAY HEAT CALCULATIONS

4.1 Comparison with experimental data and with the ENDF/B IV file

The programme gives the abundance data and their errors and, as an option, an analysis of the composition of the errors for individual nuclides in terms of contributions from uncertainties of fission yields, half-lives,average beta energies, average gamma energies, neutron capture cross-sections, and delayed-neutron branching ratios. It also gives the total decay heat and the beta and gamma contribution separately with an analysis of the composition of the errors.

The code has been used to calculate the decay heat in nuclear fuel consisting of ^{235}U choosing the conditions (irradiation and cooling times) so as to facilitate a comparison with recent integral measurements[49]. The results are shown in Figs. 1 and 2. Since the code evaluates the errors from the uncertainties in the physical input data, the uncertainties (± one standard deviation) are included in the figures in order to show the accuracy of the summation calculation. The results of summation calculations using the ENDF/B IV data file[50] are also included in the figures as solid curves (these curves are taken from ref.[49]: the calculation has been carried out by R Schenter).

In the case of the beta decay heat the agreement between the experimental values and those calculated using INVENT is excellent throughout. Nowhere in the range of cooling times covered there is any significant discrepancy. On the other hand, there are important deviations between the two summation calculations which have to be traced to either the fission yields (INVENT uses, for example,very recent experimental yields reported by Strittmatter[41]) or to the new set of average beta energies used in the present work[47].

The situation on the gamma-heat side is somewhat confusing. For cooling times above 300 s INVENT gives a slightly better fit than ENDF/B IV to the experimental data. In the range from 30 to 200 s ENDF/B IV fits the data better than INVENT, the latter giving results below the experimental ones. This situation is reversed in the range below 15 s where INVENT gives a perfect fit whereas ENDF/B IV lies below.

It is obvious that one should look for the reason why INVENT does not reproduce the experimental integral data satisfactorily over the whole range of cooling times. The recently started project aiming at measuring directly the average gamma energies of individual fission products [51] might give the answer.

At this place a discussion about the importance of the unknown fission products corresponding to about 1 % of the total fission yield is appropriate. Presumably those nuclides will have short half-lives and high decay energies which might lead to an underestimate of the decay heat during the first seconds after a reactor shut-down. It should be remembered, though, that since the cumulative yield is used for the first member of a chain only the difference between the average energy of an unknown nuclide and that of the first member considered will be missing. The uncertainty caused by the emission of unknown fission products can be, at most, a fraction of a per cent during the first seconds of cooling. It seems better to accept this small uncertainty than to include a number of hypothetical nuclides, which are completely unknown, and to estimate the effect of those on the decay heat.

Detailed comparisons with other integral decay heat determinations and with composite beta spectra are reported elsewhere [52].

4.2 Contribution from different mass regions

In order to investigate the contribution to the total decay heat from different mass regions the mass range of the fission products has been divided into six parts of roughly equal size:

I)	Left low-mass wing	A = 72 - 87
II)	Light peak	A = 88 - 102
III)	Right low-mass wing	A =103 - 117
IV)	Left high-mass wing	A =118 - 132
V)	Heavy peak	A =133 - 147
VI)	Right high-mass wing	A =148 - 166

The relative contributions from the six mass regions to the total decay power for a fuel of ^{235}U irradiated for 10^7 s are plotted in Fig. 3 as a function of cooling time. The contributions from the peaks are roughly equal and amount to about 45 % each. The wings are much less important. It might be noticed, however, that the contribution from the left high-mass wing (A = 118 - 132) exceeds 17 % at cooling times around 10^5 s.

Those mass regions which are little investigated experimentally are the right low-mass wing with contributions from the transition elements and the right high-mass wing with contributions from the rare earth elements. In these mass regions data on short-lived fission products are scarce. Fig. 3 shows, however, that these two mass regions are relatively unimportant, contributing about 3 % and 1 % respectively, to the total decay heat at cooling times below 1000 s. The scarcity of experimental results for the short-lived fission products in these mass regions is therefore not very embarrassing.

4.3 Analysis of the error of the decay power

The contributions to the error of the total decay energy in ^{235}U arising from uncertainties in the yields, capture cross sections, neutron branching ratios, decay constants, and average beta and gamma energies have been plotted versus the cooling time in Figs. 4 and 5. Fig. 4 refers

to an irradiation time of 10^7 s and Fig. 5 to an irradiation time of 1 s. These irradiation periods were chosen in order to facilitate a comparison between the uncertainties derived from the errors of the data of the individual fission products in the present approach and those obtained by Schmittroth and Schenter[9] using a more generalized sensitivity analysis.

We shall first compare the contributions from specific properties of the fission products after an irradiation time of 10^7 seconds at a flux of 1.5×10^{13} n_{th}/cm^2 s. In their treatment of the decay energies Schmittroth and Schenter assume these quantities to be correlated. Many experimental investigations of total beta decay energies have been carried out recently, supplemented by a considerable amount of detailed spectroscopic work as well as direct determinations of average beta energies. Thus, there exists now a large amount of information, and it does not seem appropriate to treat the decay energies as correlated. It has not been done in the present work, and the comparison should therefore be done with Schmittroth and Schenter's results for uncorrelated average decay energies. In Fig. 4 the contribution from average decay energies has been split up into the beta part and the gamma part. Apparently, the gamma energies are less well known giving rise to the larger contribution to the total uncertainty of the two effects, except for long cooling times ($> 10^6$ s). The sum of the beta and the gamma contributions increases from about 1,3 % at short cooling times to about 5 % at the longest time considered here. Schmittroth and Schenter give values around 1 % over the whole cooling time range, somewhat above at cooling times around 10^3 s and at very long cooling times and somewhat below at 10^5 - 10^6 s.

The reason for the increase of the contributions from the average energies with increasing cooling time is to be connected to the fact that the number of contributing fission products decreases with time and that the limits of error in the FPLIB library are conservative; the lowest error of a calculated average energy of an individual nuclide having been set equal to 10 %[47].

The per cent uncertainty arising from decay constants gradually increases from 0.7 % to 4 % in the cooling time range under study. This behaviour is again related to the decreasing number of fission products contributing. A similar behaviour has been found by Schmittroth and Schenter who, however, find lower values of the uncertainty - from less than 0.1 to about 1 %. They might somewhat underestimate the effect of the uncertainty of the decay constants in their analysis.

The contribution from the yields starts at about 0.6% at short cooling times gradually increasing to reach 3 % at 10^7 s and then decreasing somewhat. This behaviour is contrary to what is found by Schmittroth and Schenter who get an effect amounting to 0.9 % at a cooling time of 1 s decreasing to about 0.4 % at 10^8 s. The size of the effect is roughly the same as in the present work but the variation with cooling time is reversed. The reason for this is unclear. It should be pointed out, however, that FPLIB contains a large contribution of experimental yields with experimental limits of errors, especially at the low-mass peak, whereas Schmittroth and Schenter consider uncertainties in chain yields and in the parameters describing the fission charge distribution rather than the errors of the yields of the individual fission products.

The per cent uncertainties arising from the data on capture cross sections and on delayed-neutron branching ratios are small, in the range 10^{-3} - 10^{-2} in the former case and around 10^{-2} in the latter.

Finally, the total uncertainty obtained in the present work lies between 1.5 and 2.1 % for cooling times $\lesssim 10^4$s. Then it increases reaching 4% at 10^5 s and finally 7 % at 10^8 s.

A similar analysis for the case of ^{235}U irradiated for 1 s at a flux of 1.5×10^{13} n_{th}/cm^2s is shown in Fig. 5.

The contributions to the total uncertainty from decay constants, average beta energies, and average gamma energies are comparable and of the order of 1 - 3 % over a wide cooling-time range, in fair agreement with Schmittroth and Schenter's results.

For the short irradiation time the capture cross sections contribute a negligible amount to the uncertainty which is not shown in the figure.

The per cent uncertainty of the total decay heat is about 4.3 at 1 s of cooling time slowly decreasing to 2.3 at 10^3 s, then increasing again to reach 5.5 at a cooling time of 10^8 s.

As for the long irradiation period the latter increase is attributed to the decreasing number of contributing nuclides and the lower error limit 10 % used for calculated average beta and gamma energies.

Another way to investigate the errors is to plot the contribution from various sources to the per cent uncertainty of the total decay heat as a function of irradiation time for a given cooling time. This is done in Fig. 6 for zero cooling time. It is again seen that the decay constants and the average beta and gamma energies give the most important contributions to the uncertainty.

In this context the discussion about the effect of unknown and therefore omitted fission products may be taken up again. In the case of Fig. 4, which represents a long irradiation time, the nuclides contributing most to the total decay energy are quite well known, even in the cooling time range 1 - 10 s. The ten most important ones after a cooling time of 1 s are (in descending order of importance) for the beta heat: ^{92}Rb, ^{95}Sr, ^{94}Y, ^{140}Cs, ^{98}Nb, ^{139}Cs, ^{137}Xe, ^{95}Y, ^{100}Nb, and ^{92}Y. The corresponding nuclides for the gamma heat are: ^{134}I, ^{96}Y, ^{90}Rb, ^{140}La, ^{138}Cs, ^{100}Nb, ^{93}Sr, ^{142}La, ^{144}La, and ^{91}Rb. All these nuclides are known. Those cases for which the average beta decay energy has been calculated using an extrapolated beta strength function are

relatively unimportant. There the error limits are large, but the total uncertainty is little affected. For short irradiation times the importance of those nuclides will grow as is apparent from a comparison between Figs. 4 and 5, and the total uncertainty gets larger.

5. SUMMARY

A method of calculating the abundance pattern of fission products in nuclear fuel is outlined. It is especially intended to reproduce accurately the inventory of short-lived fission products. A computer code based on the method is described. A special feature is that an error analysis is included so that the uncertainty of the calculated quantities can be given directly making sensitivity checks unnecessary. The error analysis is based on the errors of the decay and cross section data and of the fission yields.

Acknowledgement

This work has been supported by the Swedish Natural Science Research Council.

Table 1

Fission products included in the library

Mass number	Isobars	References for half-lives	capture cross sections	P_n - values	Yields
71	Zn,Ga	12	36		40
72	Zn,Ga,Ge	12	36,37,39		40
73	Zn,Ga,Ge	12,13	36,39		40
74	Zn,Ga,Ge	12,14	36,37		40
75	Zn,Ga,Ge,As	12,13,14	36,37		40
76	Zn,Ga,Ge,As,Se	12,14	36,37		40
77	Zn,Ga,Ga,As,Se	12,13,14	36,39		40
78	Ga,Ge,As,Se	12,13,14	36,37,39		40
79	Ga,Ge,As,Se,Br	12,13,14,15,16	36,37,38		40
80	Ga,Ge,As,Se,Br,Kr	12,14,15	36,37		40
81	Ga,Ge,As,Se,Br	12,13,15,17,18	36,37		40
82	Ga,Ge,As,Se,Br,Kr	12,13,15	37,38		40,41
83	Ga,Ge,As,Se,Br,Kr	12,13,15,18	38		40,41
84	Ge,As,Se,Br,Kr	12,13,18,19	36,37	19	40,41
85	As,Se,Br,Kr,Rb	12,13,14,15,18	36,37,39	19	40,41
86	As,Se,Br,Kr,Rb,Sr	13,14,18,20	36,37	19	40,41
87	As,Se,Br,Kr,Rb	12,15,19,20	36,37	19	40,41,42
88	Se,Br,Kr,Rb,Sr	12,15,19,20	36,37,39	19	40,41,42
89	Se,Br,Kr,Rb,Sr,Y	12,13,15,18,20	36,37,39	19	40,41,42,43
90	Br,Kr,Rb,Sr,Y,Zr	12,13,15,21	36,38,39	19	40,41,42
91	Se,Br,Kr,Rb,Sr,Y,Zr	12,13,15,19	36,39	19	40,41,42,43
92	Br,Kr,Rb,Sr,Y,Zr	12,15,20	36,38,39	19	40,41,42,43
93	Kr,Rb,Sr,Y,Zr,Nb	12,15	36,38,39	19	40,41,42,43
94	Kr,Rb,Sr,Y,Zr,Nb,Mo	12,15,19	36,37	19	40,41,43
95	Rb,Sr,Y,Zr,Nb,Mo	12,13,14,15,19	36,39	19	40,41,43
96	Rb,Sr,Y,Zr,Nb,Mo	12,15,19,22	36,37	19	40,41
97	Rb,Sr,Y,Zr,Nb,Mo	12,13,15,19	36,38,39	19	40,41,43
98	Rb,Sr,Y,Zr,Nb,Mo	12,13,15,18,19	36,37	19	24,40,41
99	Rb,Sr,Y,Zr,Nb,Mo,Tc	12,13,19,20	36,38,39	19	24,40,41
100	Y,Zr,Nb,Mo,Tc,Ru	12,23,24	36,37		40,41
101	Zr,Nb,Mo,Tc,Ru	12	36,38,39		40,41
102	Zr,Nb,Mo,Tc,Ru	12,13,17,23	36,37		40,41
103	Nb,Mo,Tc,Ru,Rh	12,13,18,23	36,37		40,41
104	Nb,Mo,Tc,Ru,Rh,Pd	12,23	36,38,39		40,41
105	Nb,Mo,Tc,Ru,Rh,Pd	12,13,18,23	36,38,39		40,41
106	Nb,Mo,Tc,Ru,Rh,Pd	12,13,18,23	36,39		40,41
107	Mo,Tc,Ru,Rh,Pd	12,18,21	36,37,39		40
108	Mo,Tc,Ru,Rh,Pd,Ag,Cd	18,21,25	36,37		40

Mass number	Isobars	References for half-lives	capture cross sections	P_n-values	Yields
109	Tc,Ru,Rh,Pd,Ag	12,13,21,23	36,37		40
110	Tc,Ru,Rh,Pd,Ag,Cd	18,21,23,25	36,37		40
111	Ru,Rh,Pd,Ag,Cd	12,13,14,21,23,25	36,38,39		40
112	Ru,Rh,Pd,Ag,Cd	12,21	36,39		40
113	Ru,Rh,Pd,Ag,Cd,In	12,13,15,21,25	36,37,38		40
114	Rh,Pd,Ag,Cd	14,18,25	36,37		40
115	Pd,Ag,Cd,In,Sn	12,13,14,17,18	36,37,39		40
116	Pd,Ag,Cd,In,Sn	13,14	36,37		40
117	Pd,Ag,Cd,In,Sn	12,13,14,21	36,38		40
118	Pd,Ag,Cd,In,Sn	12,14,18	36,38		40
119	Ag,Cd,In,Sn	12,13,14	36,38		40
120	Ag,Cd,In,Sn	13,14,18	36,37		40
121	Ag,Cd,In,Sn,Sb	12,13,14,18	36,37		40
122	Ag,Cd,In,Sn,Sb,Te	12,14	36,37		40
123	Ag,Cd,In,Sn,Sb	12,14,26	36,37		40
124	Cd,In,Sn,Sb,Te	12,13,27	36,37		40
125	Cd,In,Sn,Sb,Te	12,13,14,28	36,37,38,39		40
126	Cd,In,Sn,Sb,Te	14,29,30	36,37,39		40
127	In,Sn,Sb,Te,I	12,13,14,26	36,38,39		40
128	In,Sn,Sb,Te,I,Xe	12,13,30	36,37		40,44,45
129	In,Sn,Sb,Te,I	12,13,14,26	36,38,39		40,45
130	In,Sn,Sb,Te,I,Xe	12,26	36,37		40,44,45
131	In,Sn,Sb,Te,I,Xe	12,13,14,26	36,38,39		40,45,46
132	In,Sn,Sb,Te,I,Xe	12,19,31,32	36,37		40,45,46
133	Sn,Sb,Te,I,Xe,Cs	12,13,14,19	36,37,38		40,45,46
134	Sn,Sb,Te,I,Xe,Cs,Ba	12,13,19,32	36,37	19	40,46
135	Sb,Te,I,Xe,Cs	12,14,18,19	36,37,38,39	19	40,46
136	Sb,Te,I,Xe,Cs,Ba	12,14,19	36,37	19	40,46
137	Te,I,Xe,Cs,Ba	12,13,19	36,38,39	19	40,42,46
138	Te,I,Xe,Cs,Ba	12,13,19	36,37	19	40,42,43,46
139	I,Xe,Cs,Ba,La	12,14,19	36,37,39	19	40,42,43,46
140	I,Xe,Cs,Ba,La,Ce	12,14,19	36,37,38,39	19	40,42,46
141	I,Xe,Cs,Ba,La,Ce,Pr	12,14	36,37,38	19	40,42,43,46
142	Xe,Cs,Ba,La,Ce,Pr,Nd	12,14,19	36,37	19	40,42,43,46
143	Xe,Cs,Ba,La,Ce,Pr,Nd	13,14,18,26	36,38,39	19	40,43,46
144	Cs,Ba,La,Ce,Pr,Nd	12,14,18,19	36,38,39	19	40,46
145	Xe,Ca,Ba,La,Ce,Pr,Nd	12,13,14,19	36,38,39	19	40,43
146	Cs,Ba,La,Ce,Pr,Nd	12,13,19	36,37	19	40
147	Cs,Ba,La,Ce,Pr,Nd,Pm,Sm	12,18,19,33	36,38,39	19	40

Mass number	Isobars	References for half-lives	capture cross sections	P_n-values	Yields
148	Ba,La,Ce,Pr,Nd,Pm,Sm	12,33	36,37		40
149	Ce,Pr,Nd,Pm,Sm	12,13,33	36,38		40
150	Ca,Pr,Nd,Pm,Sm	13,34	36,37		40
151	Pr,Nd,PmSm,Eu	12,13	36,37,38		40
152	Nd,Pm,Sm	12,35	36,37		40
153	Pm,Sm,Eu	12	36,37		40
154	Pm,Sm,Eu,Gd	12	36,37		40
155	Sm,Eu,Gd	12	36,38		40
156	Sm,Eu,Gd	12,13	36,38		40
157	Sm,Eu,Gd	12	36,38		40
158	Eu,Gd	12	36,37		40
159	Eu,Gd,Tb	12	36,38		40
160	Eu,Gd,Tb,Dy	12	36,38		40
161	Gd,Tb,Dy	12	36,37		40
162	Gd,Tb,Dy	12	36,39		40
163	Tb,Dy	12	36,39		40
164	Tb,Dy	12	36,39		40
165	Dy,Ho	18	36,37,39		40
166	Dy,Ho,Er	12	36,39		40
167	Ho,Er	12	36		40
235	U		36,39		
238	U		36,37		
239	U,Np,Pu	17	36,39		

For average beta and gamma energies ref.[47] is used throughout.

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Figure captions

Fig. 1. The quantity cooling time multiplied by beta power (in MeV/fission x s) *versus* cooling time.
Open circles with error staples (± one standard deviation): INVENT
Closed circles: Experimental results from ref.[49].
Solid curve: Summation calculation using ENDF/B IV[49,50].

Fig. 2. The quantity cooling time multiplied by gamma power (in MeV/fission x s) *versus* cooling time.
Open circles with error staples (± one standard deviation): INVENT.
Closed circles: Experimental results from ref.[49].
Solid curve: Summation calculation using ENDF/B IV[49,50].

Fig. 3. Relative contributions to the total decay heat from mass regions I - IV as functions of cooling time.
Errors correspond to ± one standard deviation.
Dotted curve: mass region 72-87.
Dash-dot curve: mass region 103-117.
Dash-dot-dot- curve: mass region 118-132.
Dashed curve: mass region 133-147.
Dash-dot-dot-dot curve: mass region 148-166.

Fig. 4. Contributions to the uncertainty of the total decay heat (gross solid curve) arising from uncertainties in yields (thin solid curve), decay constants (dotted curve), average beta and gamma energies (dashed curve and dash-dot curve), P_n-values (dash-dot-dot curve), and capture cross sections (dash-dot-dot-dot curve) as functions of cooling time. Irradiation time: 10^7 s. Note that the total effect is obtained by summing the contributions quadratically.

Fig. 5. Contributions to the uncertainty of the total decay heat (gross solid curve) arising from uncertainties in yields (thin solid curve), decay constants (dotted curve), average beta and gamma energies (dashed curve and dash-dot curve), and P_n-values (dash-dot-dot curve) as functions of cooling time. Irradiation time: 1 s. Note that the total effect is obtained by summing the contributions quadratically.

Fig. 6. Contributions to the uncertainty of the total decay heat (gross solid curve) arising from uncertainties in yields (thin solid curve), decay constants (dotted curve), and average beta and gamma energies (dashed curve and dash-dot curve) as functions of the irradiation time for cooling time zero. Note that the total effect is obtained by summing the contributions quadratically.

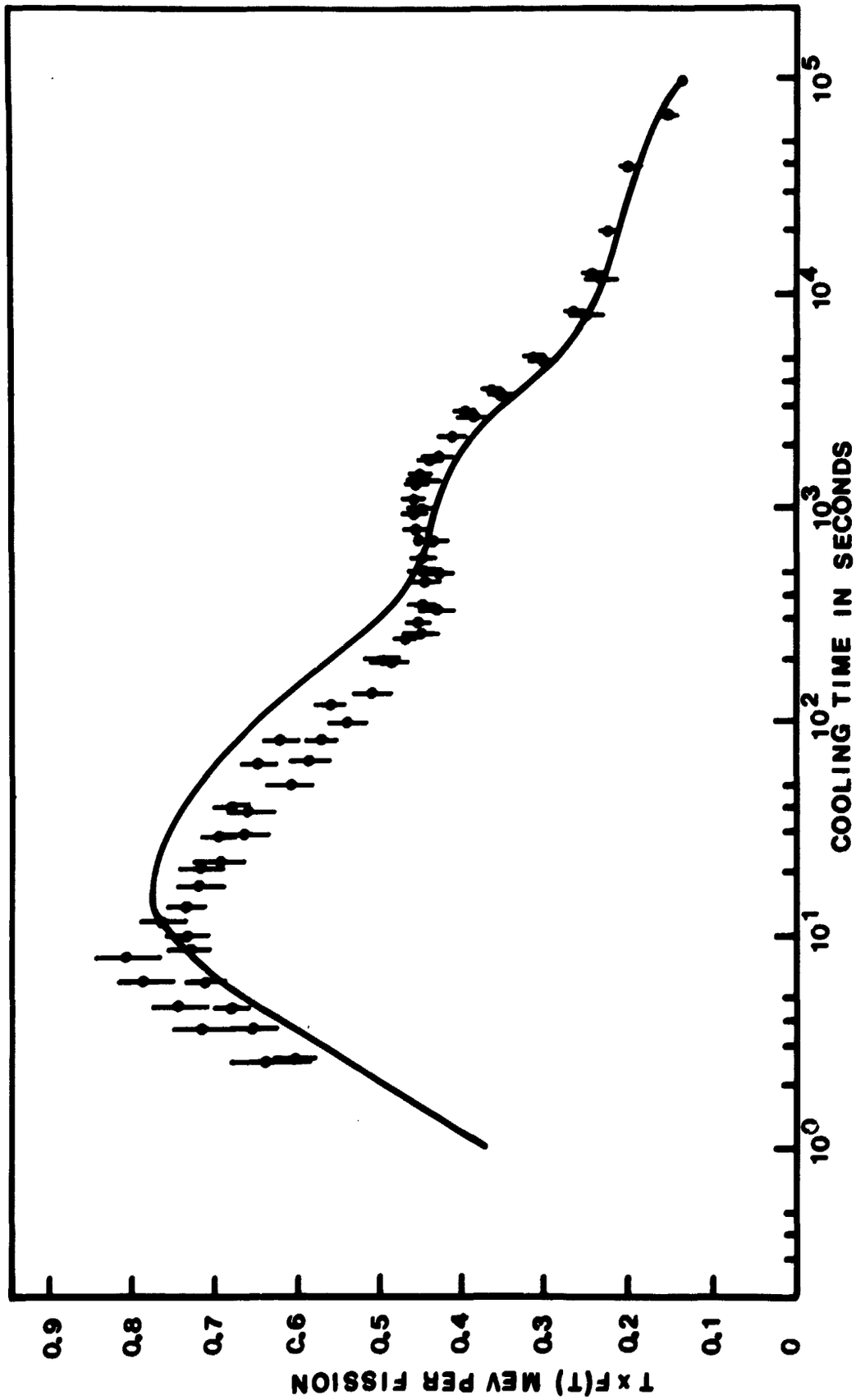
T x F(T) MEV PER FISSION
0.9
0.8
0.7
0.6
0.5
0.4
0.3
0.2
0.1
0
10^0
10^1
10^2
10^3
10^4
10^5
COOLING TIME IN SECONDS

Fig 1

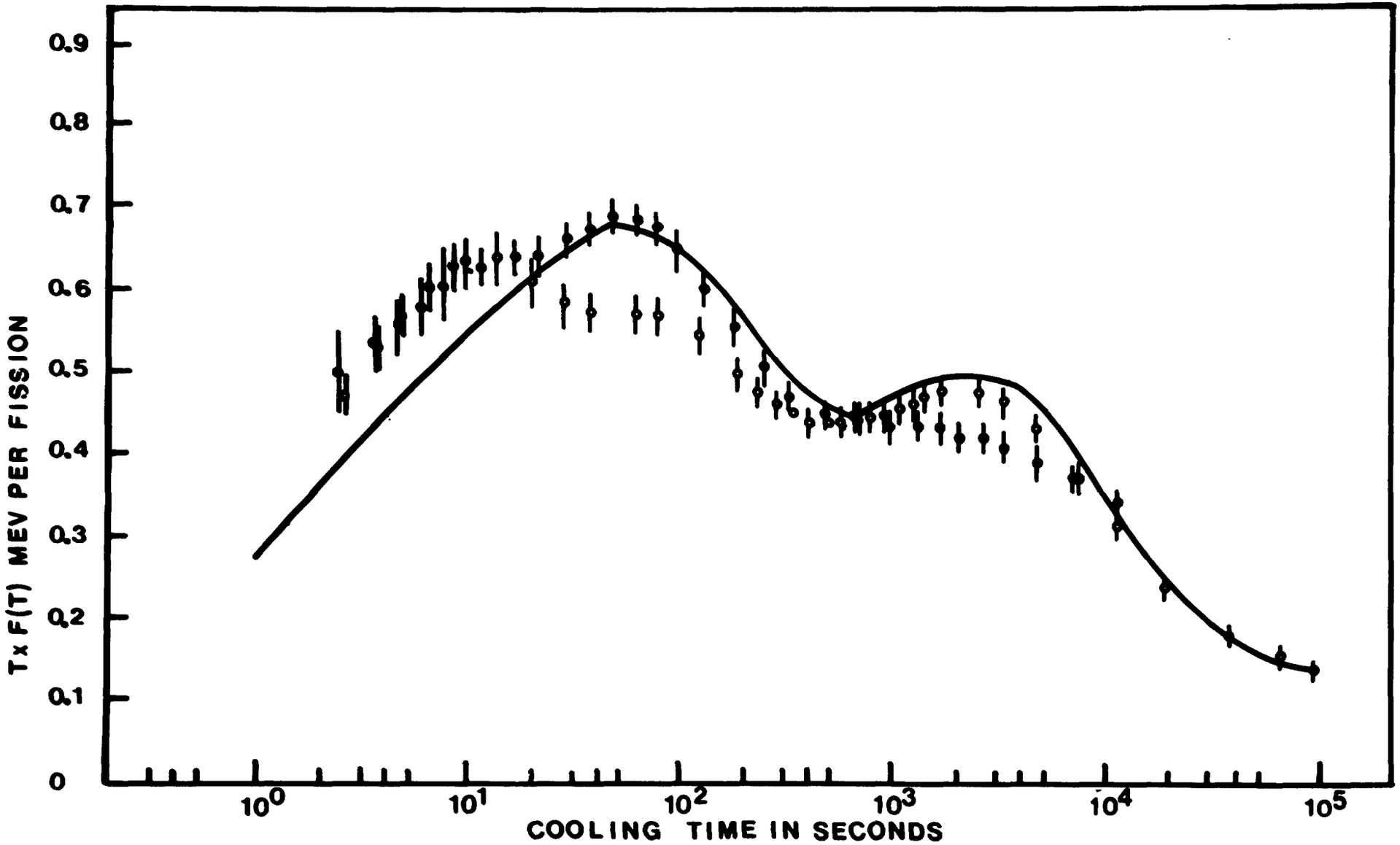
0.9
0.8
0.7
0.6
0.5
0.4
0.3
0.2
0.1
0
T x F(T) MEV PER FISSION
10^0
10^1
10^2
10^3
10^4
10^5
COOLING TIME IN SECONDS

Fig 2

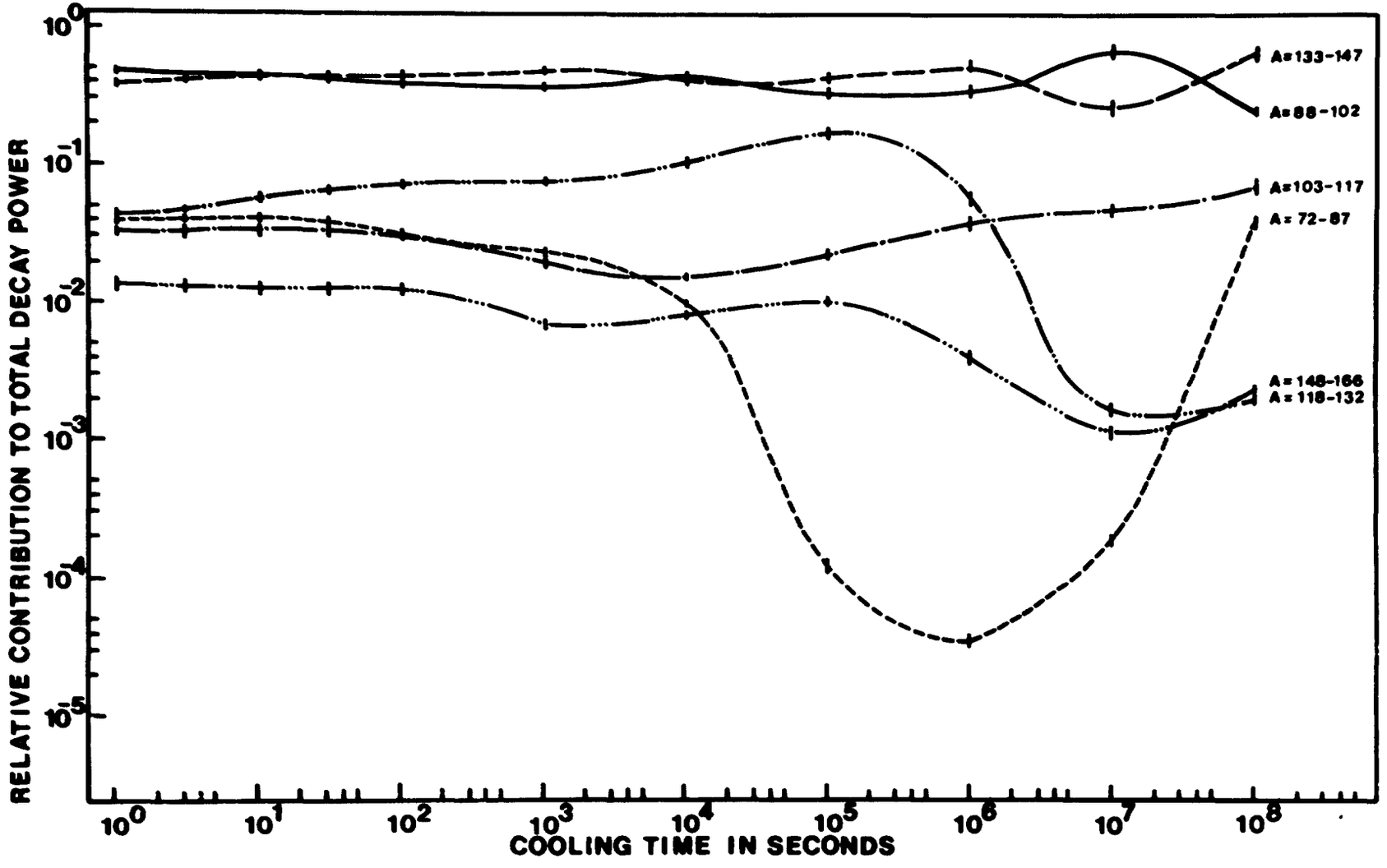
RELATIVE CONTRIBUTION TO TOTAL DECAY POWER
COOLING TIME IN SECONDS
A=133-147
A=88-102
A=103-117
A= 72-87
A=148-166
A=118-132

Fig 3

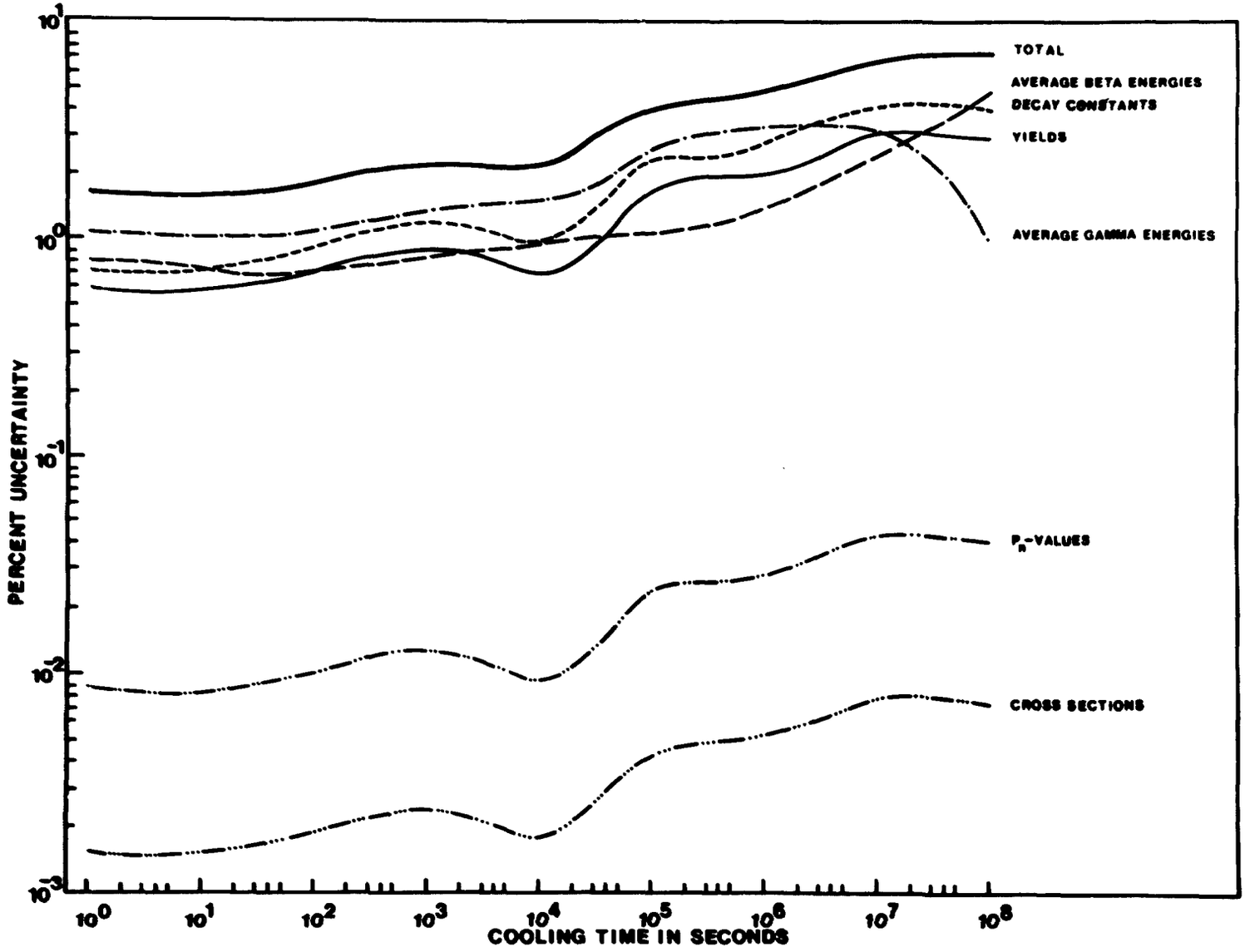
TOTAL
AVERAGE BETA ENERGIES
DECAY CONSTANTS
YIELDS
AVERAGE GAMMA ENERGIES
P_n-VALUES
CROSS SECTIONS
PERCENT UNCERTAINTY
10^1
10^0
10^{-1}
10^{-2}
10^{-3}
10^0 10^1 10^2 10^3 10^4 10^5 10^6 10^7 10^8
COOLING TIME IN SECONDS

Fig 4

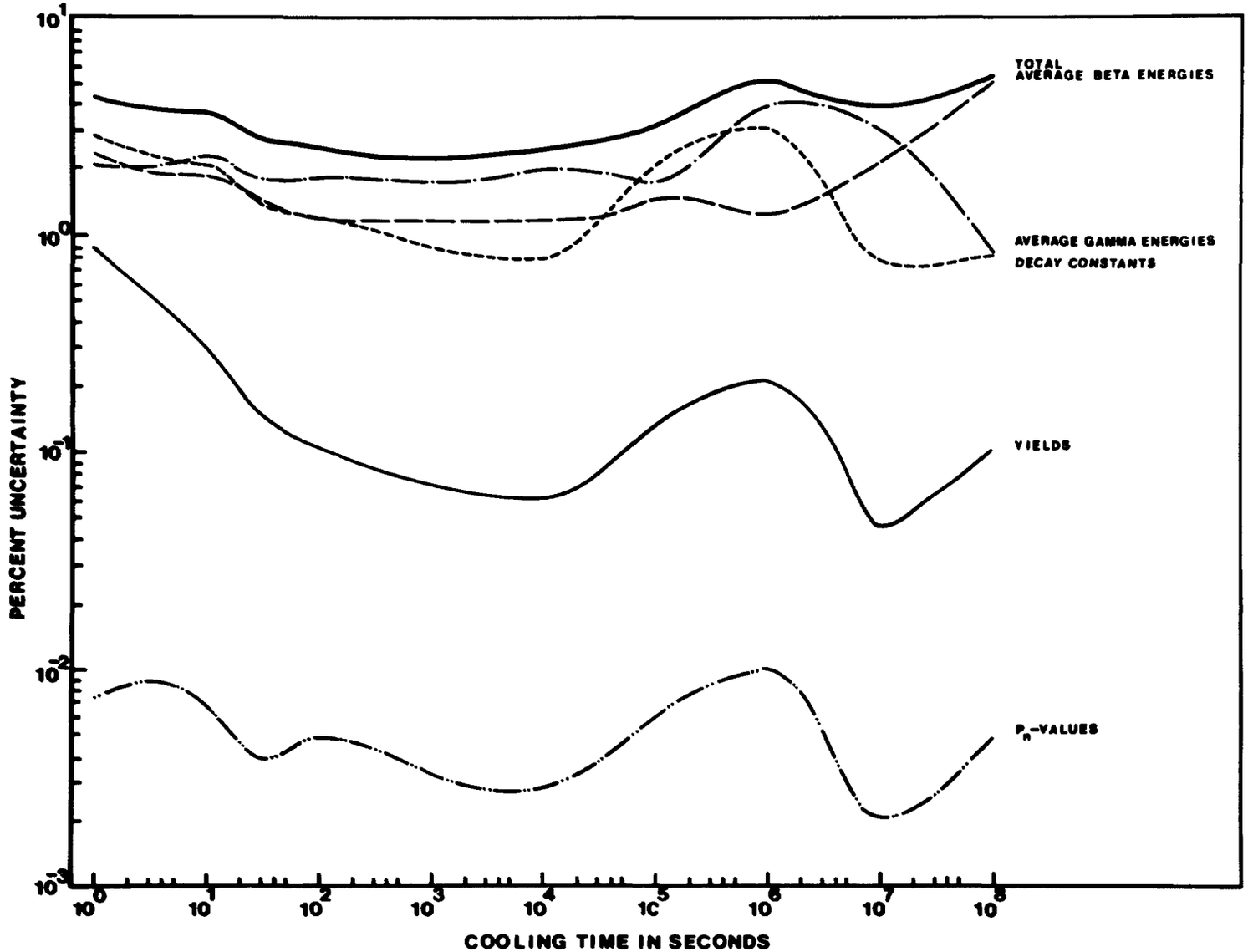
PERCENT UNCERTAINTY
COOLING TIME IN SECONDS
TOTAL AVERAGE BETA ENERGIES
AVERAGE GAMMA ENERGIES
DECAY CONSTANTS
YIELDS
P_n-VALUES

Fig 5

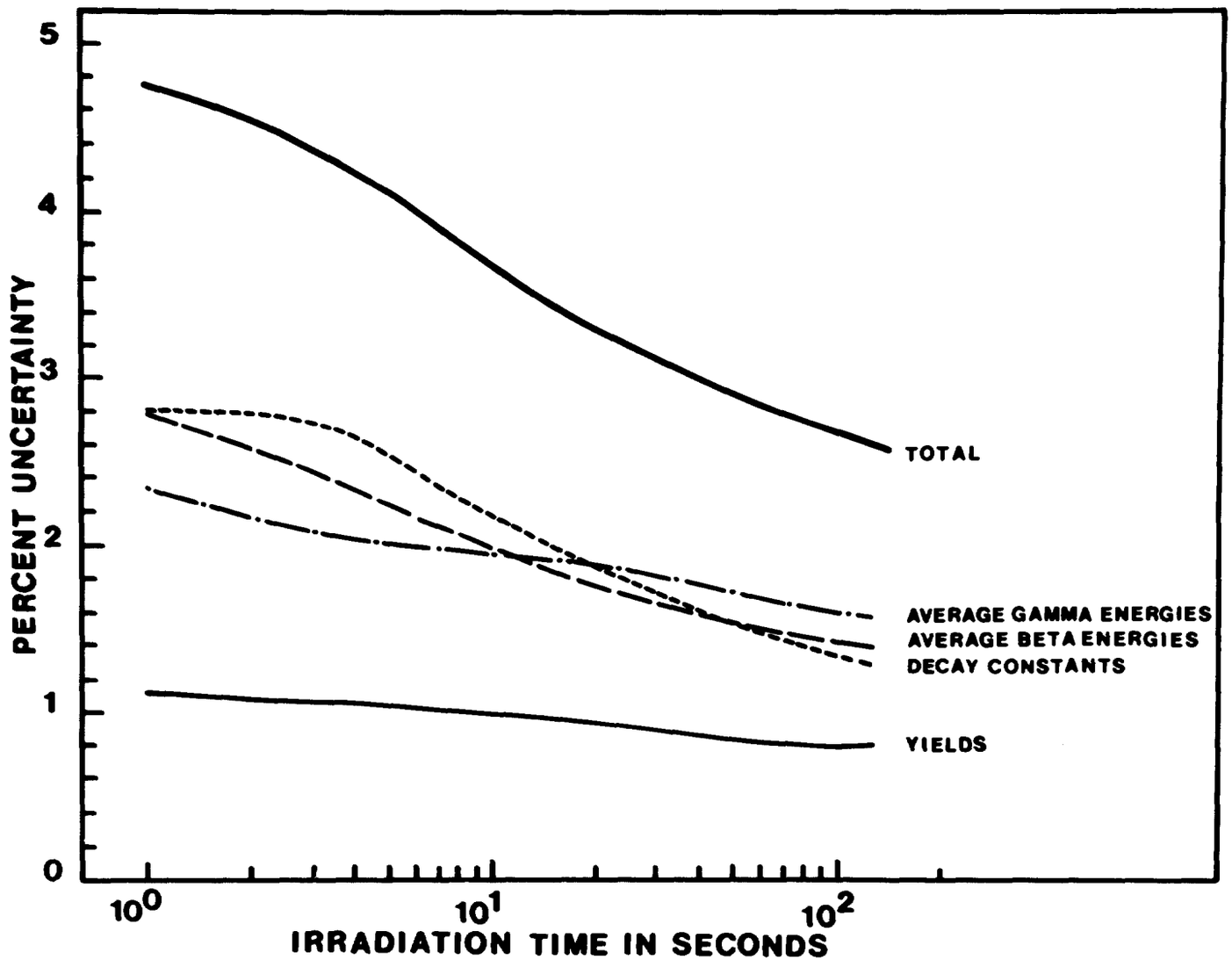
5
4
3
2
1
0
PERCENT UNCERTAINTY
TOTAL
AVERAGE GAMMA ENERGIES
AVERAGE BETA ENERGIES
DECAY CONSTANTS
YIELDS
10^0
10^1
10^2
IRRADIATION TIME IN SECONDS

Fig 6