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DETAILED COMPARISON BETWEEN DECAY HEAT DATA CALCULATED BY THE SUMMATION METHOD AND INTEGRAL MEASUREMENTS

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Detailed comparison between decay heat data calculated by the summation method and integral measurements.

by
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Abstract

The fission product library FPLIB has been used for a calculation of the decay heat effect in nuclear fuel. The results are compared with integral determinations and with results obtained using the ENDF/BIV data base. In the case of the beta part, and also for the total decay heat, the FPLIB-data seem to be superior to the ENDF/BIV-data. The experimental integral data are in many cases reproduced within the combined limits of error of the methods.

Introduction

In earlier reports the code INVENT for the evaluation of the fission product abundance pattern in nuclear fuel, and its associated data library FPLIB, have been described 1, 2). A special feature of INVENT is that it evaluates the error of the calculated quantities with account taken of the uncertainties in fission yields, half-lives, neutron cross sections, delayed-neutron branching ratios, and average beta and gamma energies of the individual fission products. This means that macroscopic properties such as the decay heat are obtained with their limits of error stated, which makes possible a proper judgement of the compatibility of calculated and experimental quantities.

The present report deals with a detailed comparison between decay heat values calculated by means of the FPLIB library and experimental integral data from beta and gamma counting and from calorimetric experiments. The calculation has been carried out using the same timing, i.e. irradiation time, waiting time, and measuring time, as the experiments in order to make the results fully comparable with experimental values. The ratio between calculated and experimental decay heat values, with the appropriate error (one standard deviation) evaluated from the errors of the nominator and the denominator, is used as a measure of the agreement between the two approaches.

As it is of great interest to compare the FPLIB-library with the ENDF/BIV-library[3)], also the ratio between values obtained using the latter data bank and the experimental values are shown, this time without error limits as those are not provided by the computer codes used in connection with ENDF/BIV.

The energy spectra of the beta particles calculated by summing the beta spectra of individual fission products using the abundances as weights have also been compared with integral measurements and with similar results obtained using ENDF/BIV.

2. Comparison with the Oak Ridge counter experiment results.

2.1 General observations

The experimental determinations by Dickens and coworkers for ^{235}U[4)] and ^{239}Pu[5)] have been chosen for a comparison with decay heat data obtained by the summation method. The experiments were carried out for three different irradiation times: 1 s, 10 s (5 s for ^{239}Pu), and 100 s. Thus three series of results have been obtained. They are plotted in Figs. 1 - 3 *versus* the average cooling time defined as the sum of the waiting time and half of the irradiation and the measuring times. The first thing to notice is that the three series of values match very well. Only rarely, as for instance in the cooling-time range 150 - 500 s for the beta heat in ^{235}U, there is a deviation amounting to more than 1 - 2 per cent.

2.2 Beta decay heat

A close examination of the beta heat results for ^{235}U (Fig. 1) shows that there are systematic trends in the data. The ratios are slightly below unity at short cooling times (below 10 s), and slightly above unity for longer cooling times. The limits of error reveal, however, that the deviations are hardly significant at any cooling time. Thus, it must be concluded that the two approaches to determine the beta part of the decay heat agree. The errors are of the same magnitude in both cases, or typically 3 - 4 %. In order to carry the comparison to a higher precision it is therefore necessary to improve both the experimental integral determinations and the basic data, especially the average beta and gamma energies, of the individual nuclides entering into the FPLIB-library.

The ENDF/BIV-results do not reproduce the experimental values as well as FPLIB. This holds both for short cooling times where they are too low and for the intermediate cooling time range 10 - 100 s where they are too high. For longer cooling times both summation treatments are comparable in precision. In the absence of error estimates it is not possible to judge whether there is a significant disagreement between ENDF/BIV and experiments, however.

For ^{239}Pu the situation is much the same as for ^{235}U. The beta ratios are below unity for cooling times shorter than about 20 s and then above. The deviation - about one standard deviation except for the shortest cooling times where it amounts to about two standard deviations - is again so small than one must consider the two approaches to agree within limits of error.

Also for the ENDF/BIV-file the results for ^{239}Pu are similar to those for ^{235}U. Again, the FPLIB-file seems to be superior to the ENDF/BIV-file below about 500 s.

2.3 Gamma decay heat

Turning now to the gamma decay heat one notices for ^{235}U a good agreement between FPLIB and experiment below about 20 s of cooling and above about 200 s (of Fig. 2). In the intermediate range the agreement is poor, and there seems to be a significant difference between the summation method and the experimental data. Thus, the set of gamma energies in the FPLIB-file must be checked carefully. To this end an experimental project has been started at this laboratory aiming at determining directly the average gamma energies of short-lived fission products[6].

When compared to the ENDF/BIV results the FPLIB-based calculation seems to be superior for short cooling times whereas ENDF/BIV gives results in better agreement with experiments in the range 20 - 300 s. For still longer cooling times FPLIB is again the better choice.

The ^{239}Pu-results for the gamma heat are similar to the ^{235}U-ones. There is agreement within limits of error for short and long cooling times but not in an intermediate cooling time range which now extends from about 300 to about 3000 s. When compared to ENDF/BIV, FPLIB is superior for short cooling times and ENDF/BIV gives better agreement in an intermediate region up to about 500 s. After that, both summation calculations give very similar results.

2.4 Total decay heat

The results for the total decay heat are shown in Fig. 3. The low gamma results at intermediate cooling times are now compensated by beta results slightly above unity in the same cooling time range. Although the ratios for both ^{235}U and ^{239}Pu are slightly below unity (except for ^{235}U and long cooling times) the deviations are hardly of a significant magnitude. In comparison with ENDF/BIV the FPLIB-file seems to be superior except for long cooling times where both summation calculations give very similar results.

3. Comparison with the Los Alamos calorimetric experiment results

The comparisons have so far been done with experimental results obtained for rather short irradiation and cooling times. It is therefore of interest to compare the summation method with integral experimental data for other conditions, namely with the set of data determined by Yarnell and Bendt using a calorimetric method [7,8]. In their experiments the irradiation time was 20 000 s, and the cooling time was varied from 10 s to 100 000 s.

The decay heat results are shown in Fig. 4. For ^{235}U the agreement is very good except for short cooling times where the ratio differs from unity by about two standard deviations. These differences are hardly significant and it must be concluded that the methods of approach agree within their limits of error. It should be noted that ENDF/BIV is in better agreement with experiments at the shortest cooling times. For longer times the two summation calculations agree.

In the case of ^{239}Pu the ratios are systematically below unity. The limits of error are larger here but, at least for cooling times below some 500 s, the difference seems to be significant. The ENDF/BIV-calculation and the FPLIB-calculation agree with each other over the whole range of cooling times.

4. Comparison with beta energy spectra

In their experiment on the beta part of the decay heat Dickens et al 4) determined the energy spectra of the beta particles for various irradiation and cooling times. The corresponding spectra can also be obtained by summing the spectra of individual fission products obtained in the work reported in ref. 9), properly weighted according to their abundances as determined with the code INVENT. In order to facilitate a comparison also with results using ENDF/BIV (from ref. 10)) the spectra shown in this section have been plotted in units of MeV per fission and bin (number of beta particles per second and bin (of magnitude 100 keV) multiplied by the energy of the bin (in MeV) and divided by the number of fissions per second during the irradiation period).

In Figs. 5 - 7 experimental results for ^{235}U are shown as a hatched area (corresponding to ± one standard deviation), and the present results of the calculation are given as solid curves. The broken curves correspond to a summation calculation using the ENDF/BIV data base and are taken from ref. 10).

Energy spectra obtained at cooling times from 2.2 to 17.2 s after an irradiation time of 1 s are shown in Fig. 5. The agreement between calculation and experiment is excellent at all these cooling times, and no significant deviations are found anywhere in the spectra.

It is also evident that the FPLIB-calculation agrees better than the ENDF/BIV one, especially at the low-energy end of the spectra.

The spectra at intermediate cooling times, from 20.7 s to 254.7 s, and an irradiation time of 10 s are given in Fig. 6. The agreement is also here good at the shorter and the longer cooling times.At the intermediate cooling times, 39.7 s and 84.1 s, however, the calculated spectra tend to lie above the experimental ones at energies above about 3 MeV. In this range ENDF/BIV gives a better fit whereas the FPLIB-calculation reproduces the experimental data better at lower energies.

The situation at long cooling times, from 450 s to 4950 s, and for an irradiation time of 100 s, is again very good. The calculation reproduces the experimental spectra well at all the cooling times. The FPLIB- and the ENDF/BIV-libraries yield almost identical results at this cooling time range.

5. Discussion

The detailed comparisons carried out in the present report show that the beta part of the decay heat, when calculated by the summation method using the FPLIB-data base, are in as good agreement with experimental data as can be expected from the combined limits of error. The errors of both methods of approach are of the same order of magnitude and, consequently, to carry the comparison further requires improving both the accuracy of the experimental data and of the physical data forming the base for the calculation.

In comparison with the ENDF/BIV data base the FPLIB-library seems to be the better choice at short to intermediate cooling times. At long cooling times both libraries yield similar results.

In the case of the gamma part of the decay heat there is a disagreement at intermediate cooling times which indicates that the basic data, especially the average gamma energies of those nuclides which are the more important contributors in this cooling time range, should be studied carefully.

For the total energy release the calculation using FPLIB yields results in agreement with experiment over all cooling times checked here.

Finally, with few exceptions, the calculated beta spectra reproduce the measured ones very well.

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Captions of figures

Fig. 1. Ratio between beta decay heat values calculated by means of the FPLIB-library and experimental data from refs.4) and 5): (a) ^{235}U, (b) ^{239}Pu. The errors correspond to ± one standard deviation.

Open circles: Irradiation time 1 s.

Closed circles: Irradiation time 10 s for ^{235}U and 5 s for ^{239}Pu.

Open triangles: Irradiation time 100 s.

Ratios obtained using the ENDF/BIV data base for evaluating the decay heat are indicated by broken curves, dotted curves and dash-dot curves for the irradiation times 1 s, 10 (5) s, and 100 s, respectively.

Fig. 2. Ratio between gamma decay heat values calculated by means of the FPLIB-library and experimental data from refs.4) and 5): (a) ^{235}U, (b) ^{239}Pu. The errors correspond to ± one standard deviation.

Open circles: Irradiation time 1 s.

Closed circles: Irradiation time 10 s for ^{235}U and 5 s for ^{239}Pu.

Open triangles: Irradiation time 100 s.
Ratios obtained using the ENDF/BIV data base for evaluating the decay heat are indicated by broken curves, dotted curves and dash-dot curves for the irradiation times 1 s, 10 (5) s, and 100 s, respectively.

Fig. 3. Ratio between total decay heat values calculated by means of the FPLIB-library and experimental data from refs.4) and 5): (a) ^{235}U, (b) ^{239}Pu. The errors correspond to ± one standard deviation.

Open circles: Irradiation time 1 s.

Closed circles: Irradiation time 10 s for ^{235}U and 5 s for ^{239}Pu.

Open triangles: Irradiation time 100 s.

Ratios obtained using the ENDF/BIV data base for evaluating the decay heat are indicated by broken curves, dotted curves and dash-dot curves for the irradiation times 1 s, 10 (5) s, and 100 s, respectively.

Fig. 4. Ratio between total decay heat values calculated by means of the FPLIB library and experimental data from refs.[7] and [8]: (a) ^{235}U, (b) ^{239}Pu. The errors correspond to ± one standard deviation. Ratios obtained using the ENDF/BIV data base for evaluating the decay heat are indicated by broken curves.

Fig. 5. Beta energy release in ^{235}U in units of MeV per fission and energy bin of size 100 keV for an irradiation time of 1 s.

(a) Cooling time 2.2 s, (b) Cooling time 4.2 s,
(c) Cooling time 8.2 s, (d) Cooling time 17.2 s.

Hatched areas: Experimental data from ref. [4] (taken from ref.[10]) with area corresponding to ± one standard deviation.

Solid curves: Spectra calculated by means of the FPLIB library.

Broken curves (shown only if different from the solid ones): Spectra calculated by means of the ENDF/BIV library (from ref. [10]).

Fig. 6. Beta energy release in ^{235}U in units of MeV per fission and energy bin of size 100 keV for an irradiation time of 10 s.

(a) Cooling time 20.7 s, (b) Cooling time 39.7 s,
(c) Cooling time 84.7 s, (d) Cooling time 254.7 s.

Hatched areas: Experimental data from ref. 4) (taken from ref. 10)) with area corresponding to ± one standard deviation.

Solid curves: Spectra calculated by means of the FPLIB library.

Broken curves (shown only if different from the solid ones): Spectra calculated by means of the ENDF/BIV library (from ref. 10)).

Fig. 7. Beta energy release in ^{235}U in units of MeV per fission and energy bin of size 100 keV for an irradiation time of 100 s.

(a) Cooling time 450 s, (b) Cooling time 950 s, (c) Cooling time 2700 s, (d) Cooling time 4950 s.

Hatched areas: Experimental data from ref. 4) (taken from ref. 10)) with area corresponding to ± one standard deviation.

Solid curves: Spectra calculated by means of the FPLIB library.

Broken curves (shown only if different from the solid ones): Spectra calculated by means of the ENDF/BIV library (from ref. 10)).

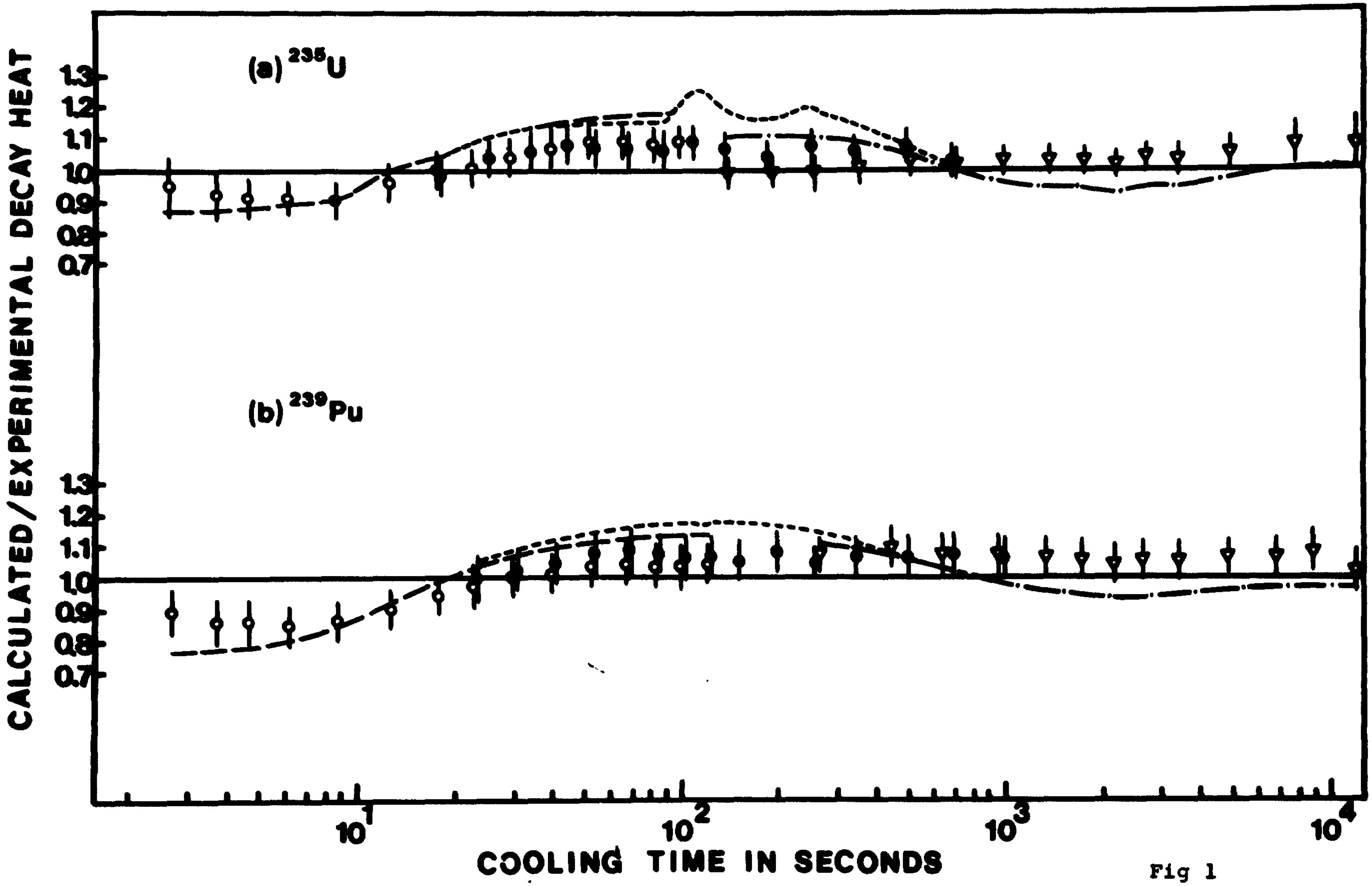
(a) ^{235}U
(b) ^{239}Pu
CALCULATED/EXPERIMENTAL DECAY HEAT
1.3
1.2
1.1
1.0
0.9
0.8
0.7
10^1
10^2
10^3
10^4
COOLING TIME IN SECONDS

Fig 1

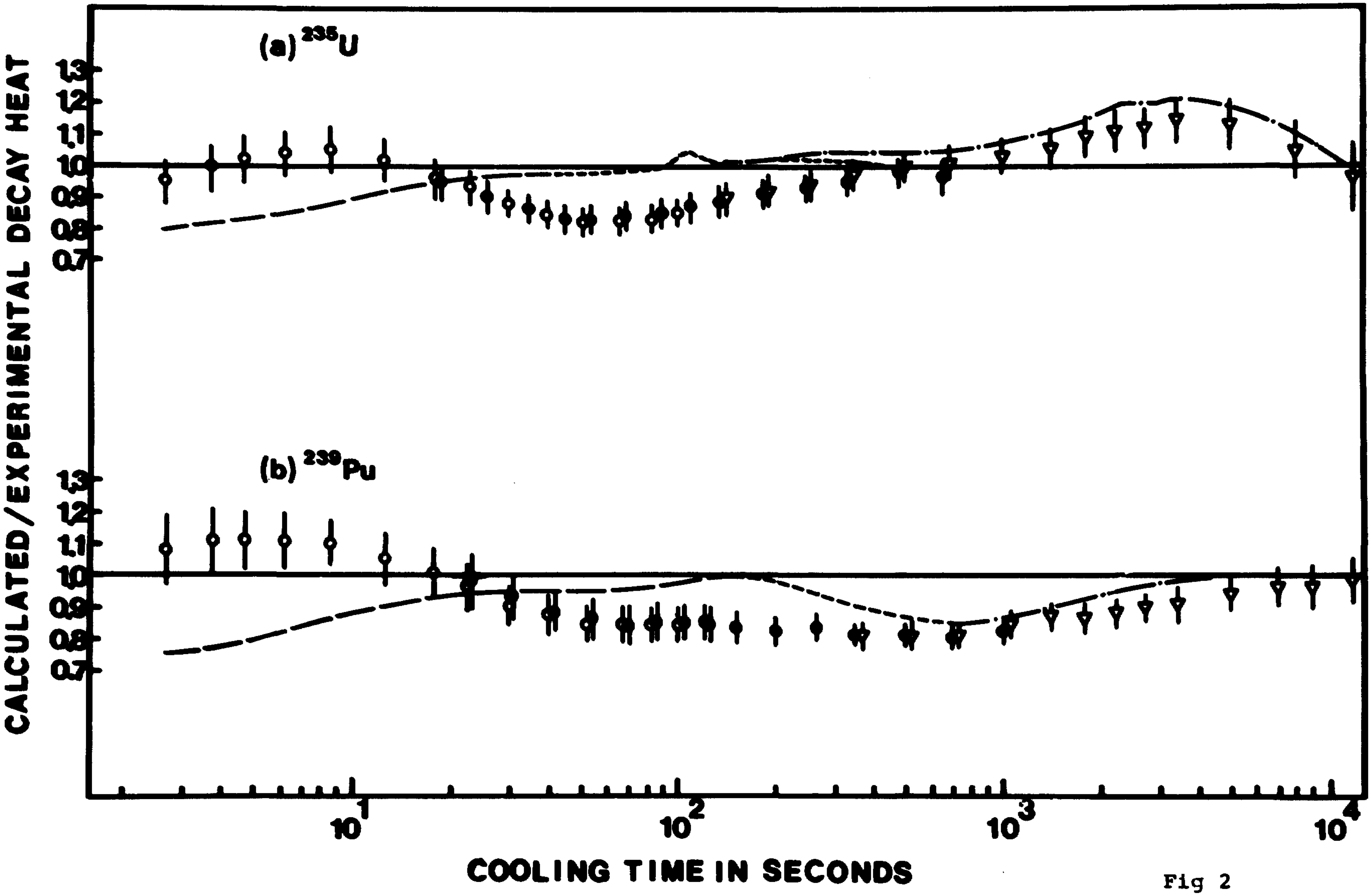
(a) ^{235}U
(b) ^{239}Pu
1.3
1.2
1.1
1.0
0.9
0.8
0.7
CALCULATED/EXPERIMENTAL DECAY HEAT
10^1
10^2
10^3
10^4
COOLING TIME IN SECONDS

Fig 2

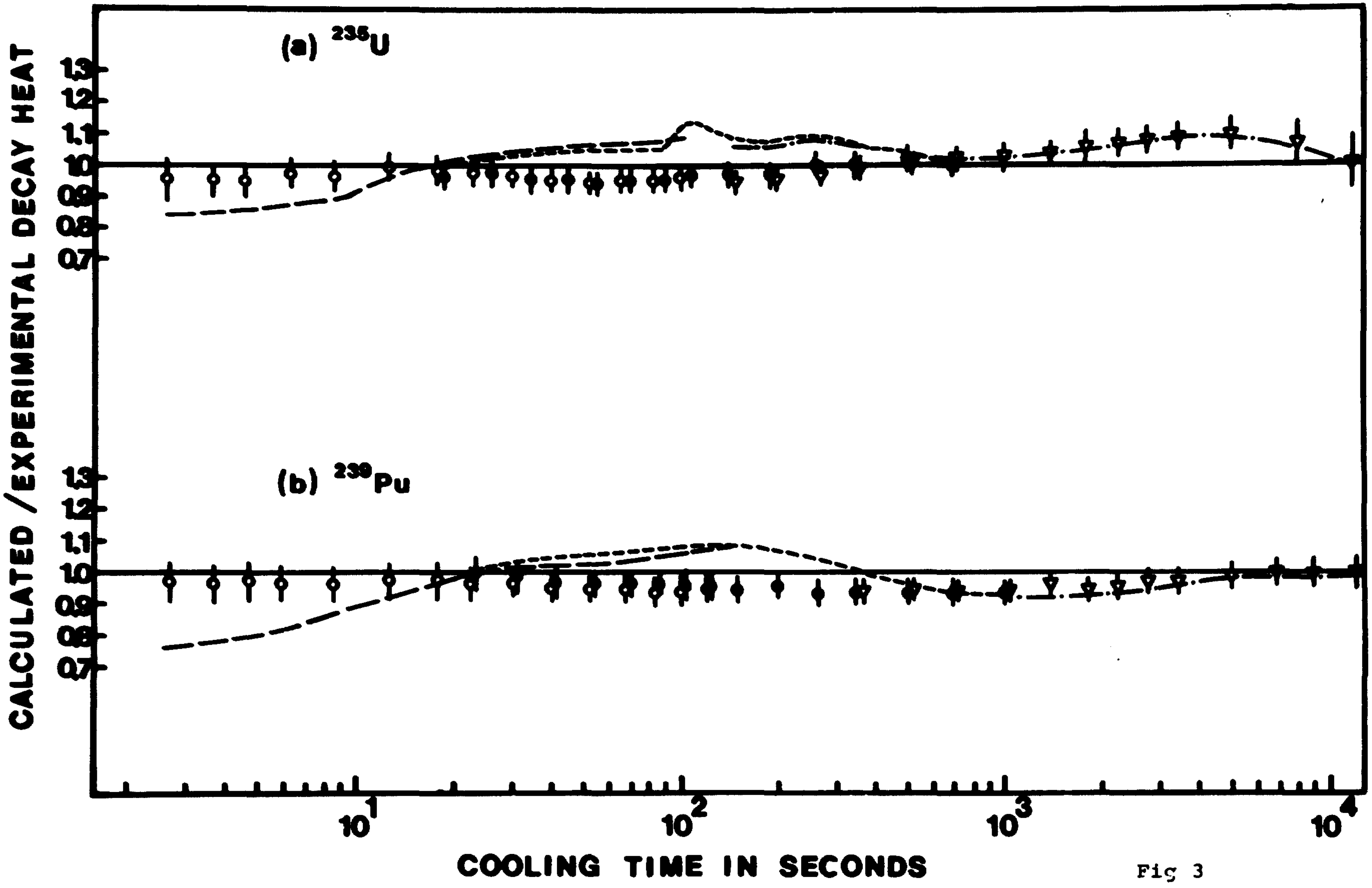
(a) ^{235}U
(b) ^{239}Pu
CALCULATED / EXPERIMENTAL DECAY HEAT
1.3
1.2
1.1
1.0
0.9
0.8
0.7
10^1
10^2
10^3
10^4
COOLING TIME IN SECONDS

Fig 3

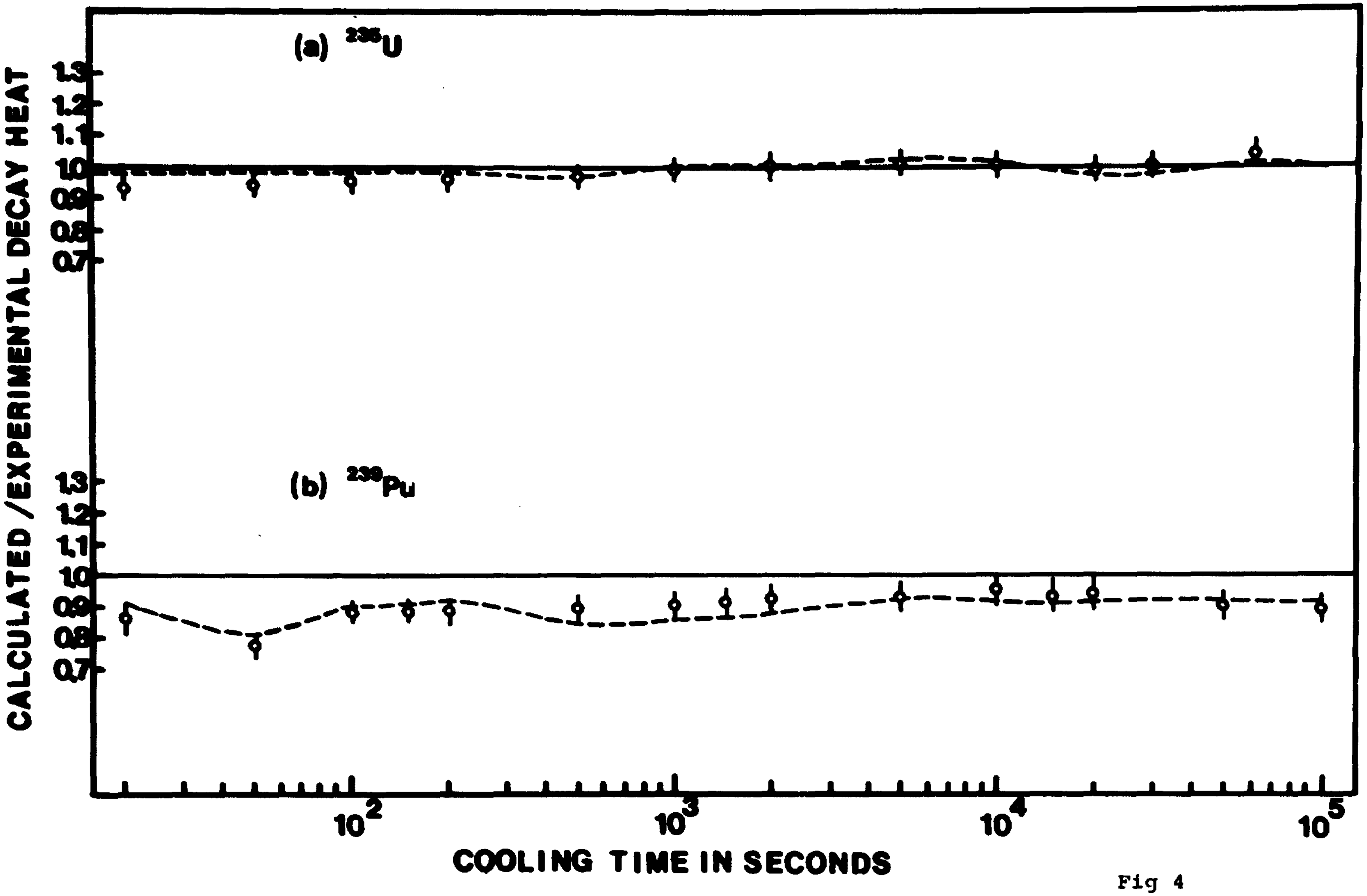
(a) ^{235}U
(b) ^{239}Pu
1.3
1.2
1.1
1.0
0.9
0.8
0.7
CALCULATED /EXPERIMENTAL DECAY HEAT
10^2
10^3
10^4
10^5
COOLING TIME IN SECONDS

Fig 4

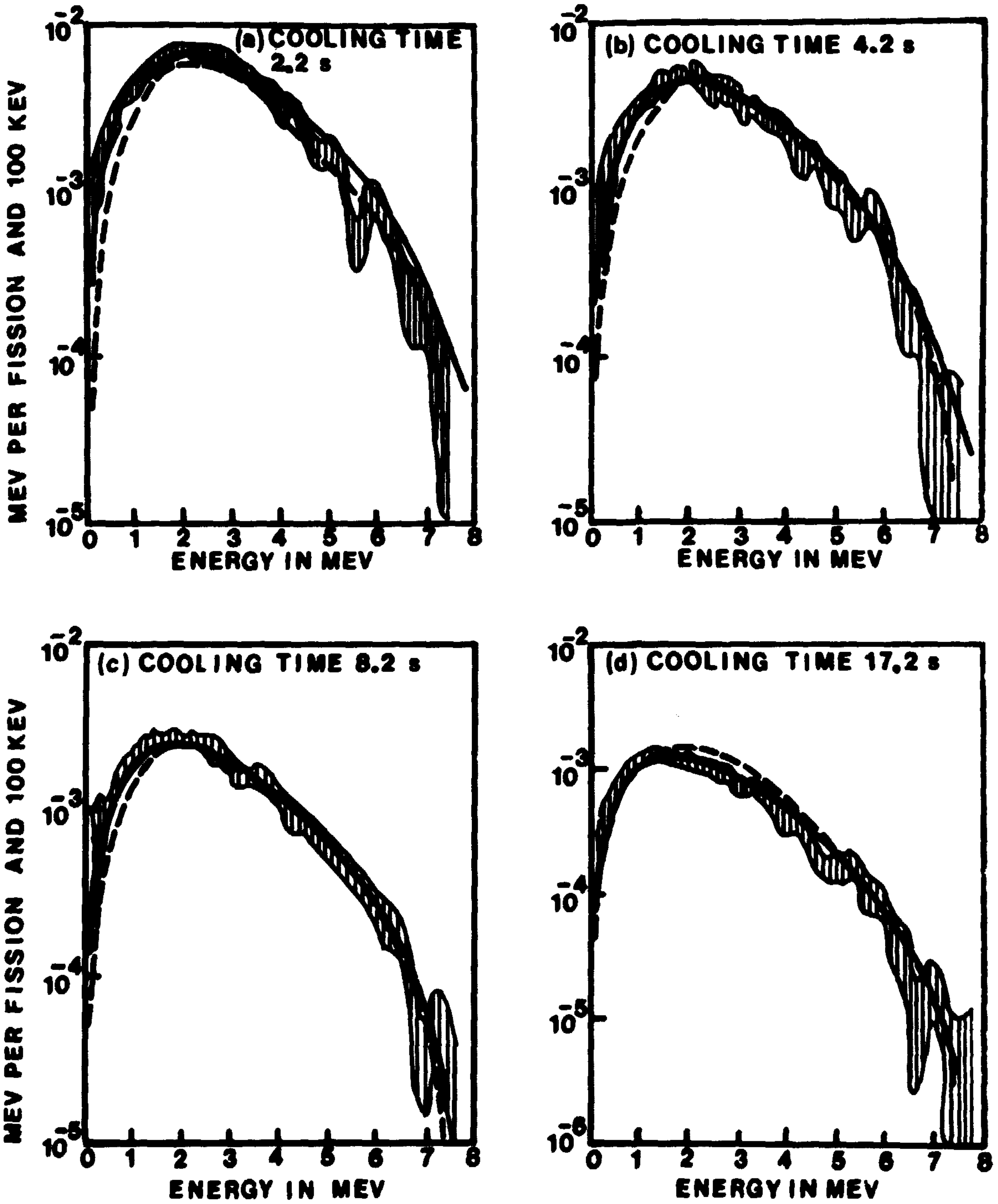
(a) COOLING TIME 2.2 s
(b) COOLING TIME 4.2 s
(c) COOLING TIME 8.2 s
(d) COOLING TIME 17.2 s
MEV PER FISSION AND 100 KEV
ENERGY IN MEV
10^{-2}
10^{-3}
10^{-4}
10^{-5}
10^{-6}
0
1
2
3
4
5
6
7
8

Fig 5

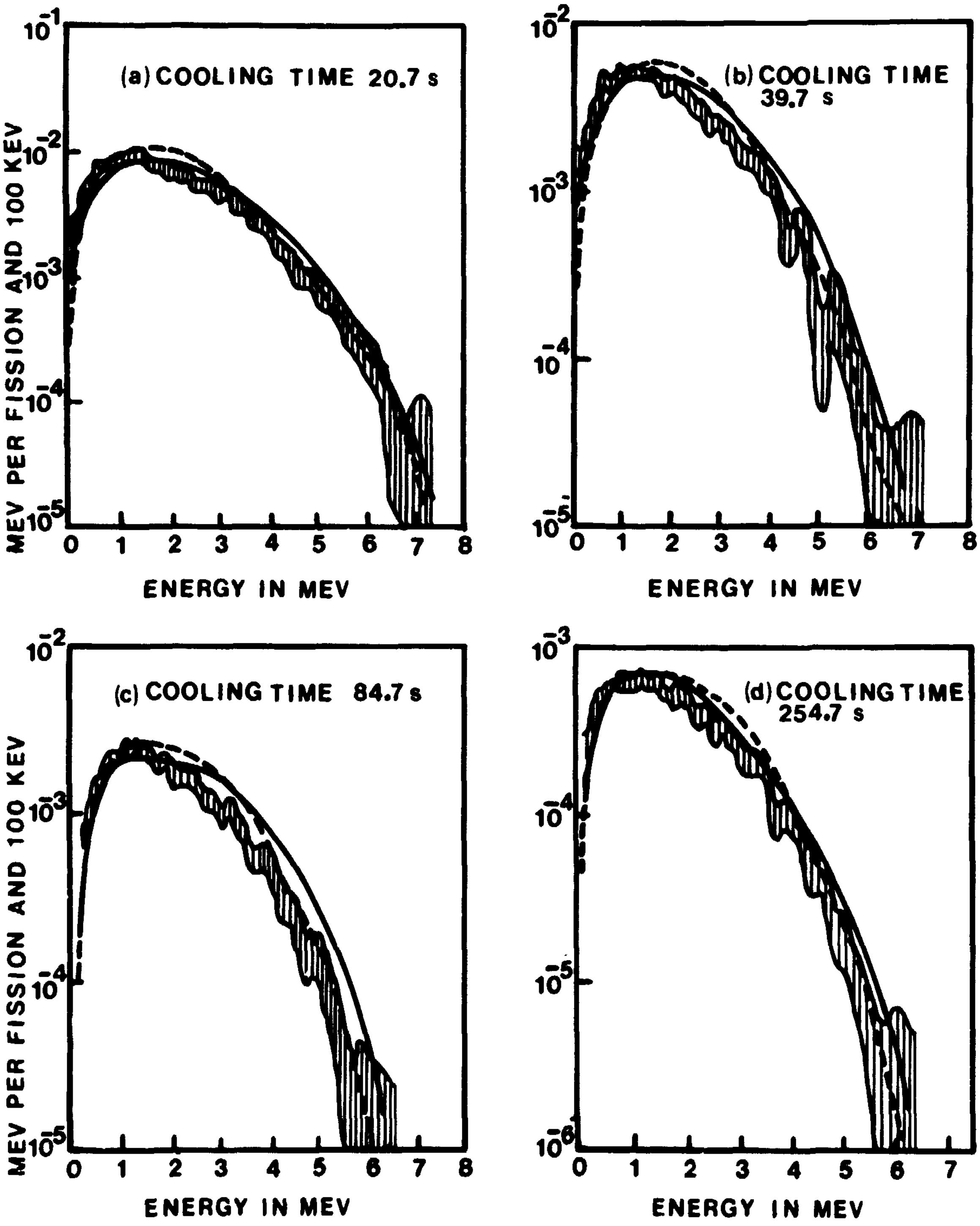
(a) COOLING TIME 20.7 s
(b) COOLING TIME 39.7 s
(c) COOLING TIME 84.7 s
(d) COOLING TIME 254.7 s
MEV PER FISSION AND 100 KEV
ENERGY IN MEV
10^{-1}
10^{-2}
10^{-3}
10^{-4}
10^{-5}
10^{-6}
0
1
2
3
4
5
6
7
8

Fig 6

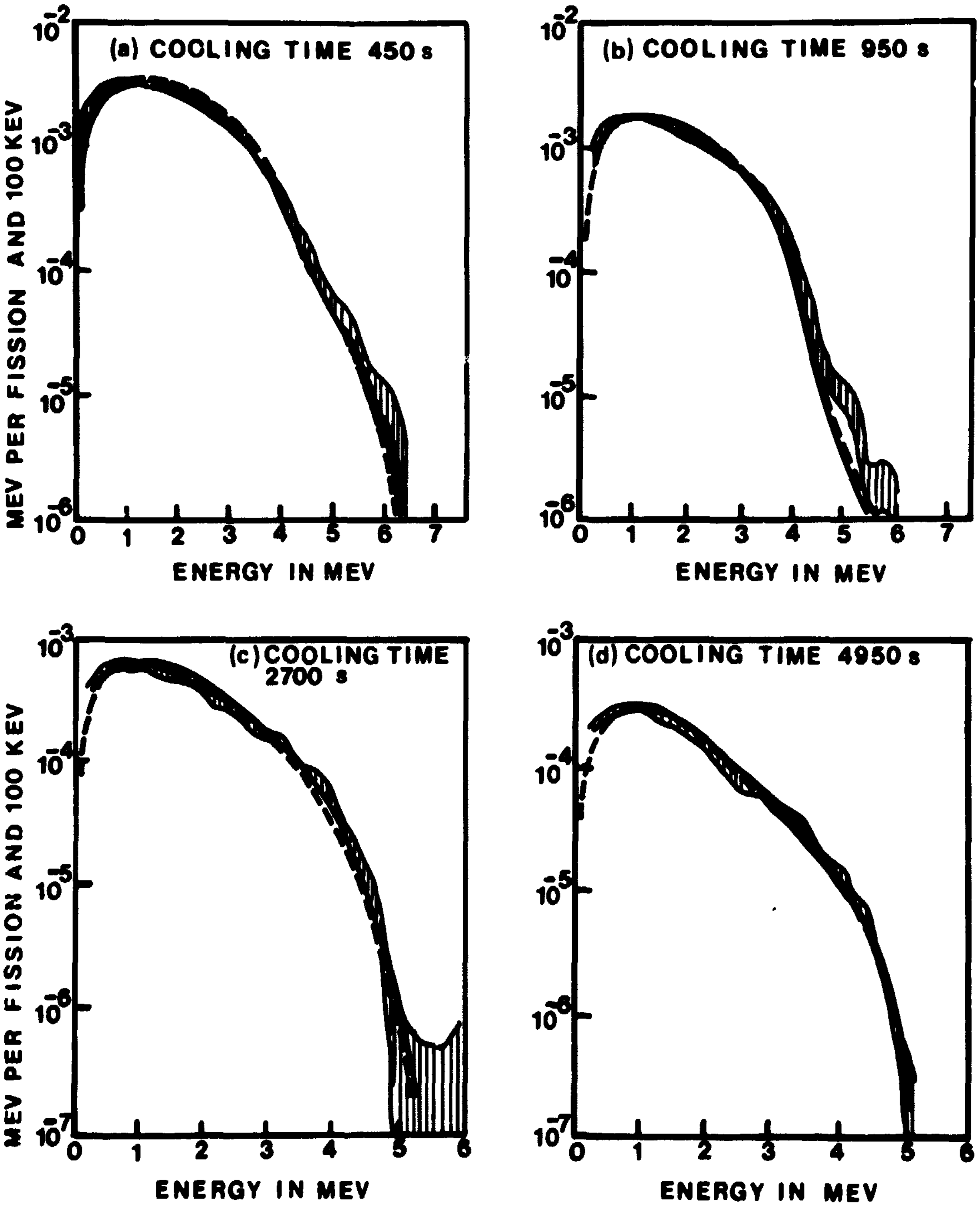
(a) COOLING TIME 450 s
(b) COOLING TIME 950 s
(c) COOLING TIME 2700 s
(d) COOLING TIME 4950 s
MEV PER FISSION AND 100 KEV
ENERGY IN MEV
10^{-2}
10^{-3}
10^{-4}
10^{-5}
10^{-6}
10^{-7}
0
1
2
3
4
5
6
7

Fig 7