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Decay Heat Predictions Using JEF1

- by -

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For the attention of: the UK Chemical Nuclear Data Committee,
the Differential and Integral Data Study Group and
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SUMMARY

The first Joint Evaluated File (JEF1) of data for reactor calculations has been constructed under the auspices of the NEA Data Bank. The data available within JEF1 for the calculation of decay heat due to direct fission products has been examined and the evaluation procedures used to produce these data are described.

Decay heat predictions using the JEF1 data have been compared with corresponding values obtained with the UK data files. Differences of up to a few percent are observed in the predictions for a fission pulse. These occur mainly at short cooling times and can be attributed to revised fission yield data. For practical applications the differences in integral predictions using JEF1 and UK data are shown to be much smaller. As a consequence of improved data for short lived fission products in JEF1, predicted gamma spectra at short cooling times will be more complete than those obtained with the UK data. It is concluded that for total decay heat predictions there is little to choose between UK data and that of JEF1. However, for applications which require spectral predictions at short cooling times the use of JEF1 is preferred.

UK and JEF1 total decay heat predictions have also been compared with results of a least squares fit to measured data for both U235 and Pu239 and directly with results of measurements for U238 and Pu241. Acknowledged deficiencies in decay data for short lived fission products (half lives < 100 s) are confirmed by the observed differences between measurement and prediction at short cooling times. However, it was found that both UK and JEF1 predictions display a discrepancy of 5-10% with repect to measurement for cooling times around 1000 s when the decay data were considered reliable. Since this discrepancy may lead to uncertainties which exceed the accuracy requirements for some applications (typically 5 %) it is necessary to identify its cause and seek suitable improvements to the basic data.

The principal decay heat nuclides at a cooling time of 1000 s were identified and it was found that, for a number of them, the decay characteristics had been determined from relatively few measurements. It was also shown that the decay data for these nuclides, present in both UK and JEF1 data libraries, offered scope for revision which would permit improvement in the agreement between measurment and prediction.

As plans for JEF2 are prepared, consideration should be given to improve decay heat predictions. It has been shown that while there is a general requirement for further data on short lived nuclides there is also a specific need to re-examine experimentally the decay schemes of a number of fission products with effective half lives of around 1000 s.

1.0 INTRODUCTION

Decay heat is the term applied to the heat generation within the fuel of fission reactors after irradiation has ceased. It arises principally from the beta and gamma radiation released in the radioactive decay of the fission products produced in the fission reaction. There is also a contribution from the alpha, beta and gamma radiation emitted in the radioactive decay of heavy elements which arise from the transmutation of the fuel materials. An accurate knowledge of this decay heat is required for reactor fault studies while details of the gamma energy release are needed in the assessment of shielding requirements for fuel discharge, storage and transportation, and for the long term storage of radioactive waste. Table 1 summarises the accuracy requirements for decay heat estimates in different reactor systems as well as for fuel handling and storage. A comprehensive review covering all aspects of decay heat analysis is given by Tobias (1980).

This note examines the nuclear data requirements for decay heat evaluation, in particular the fission yield and decay data for fission products. The data available in the first Joint Evaluated File (JEF1) are summarised and the results of decay heat calculations using these data are presented. By comparing results of decay heat measurements with these and other predictions an attempt is made to identify areas where further improvements in basic data are required.

2.0 BASIC CONCEPTS IN DECAY HEAT EVALUATION

Before proceeding further it is desirable to introduce two basic concepts which are used widely in the field of decay heat analysis. The fission pulse function at a cooling time t (for any fissile species) is defined as the mean decay energy release rate per unit fission at that time following an instantaneous pulse of many fissions. For the present purposes this will be denoted by f(t). Conventional units for the fission pulse function are MeV/f/s. Since f(t) varies approximately as 1/t it is common practice to exhibit decay heat fission pulse data in the form f(t).t versus t.

If neutron capture effects in fission products are ignored then an extended irradiation of duration I, at constant fission rate, can be considered to be equivalent to a succession of fission pulse functions each scaled by the fission rate. It follows that the energy release rate per unit fission rate, at a time t following the end of irradiation, is given by the integral of the pulse function between the limits t and I+t. Thus, if this is represented by F(I,t) then

$$F(I,t) = \int_{t}^{I+t} f(t)\,dt$$

It follows from the basic properties of integrals that the right hand side of this equation may be reduced to a number of integrals covering consecutive time intervals. For some arbitrary time T the above equation becomes

$$F(I,t) = \int_t^T f(t)\,dt + \int_T^{I+t} f(t)\,dt$$

thus

$$F(I,t) = \int_t^T f(t)\,dt - \int_{I+t}^T f(t)\,dt$$

If T is very much greater than both I and t it can be considered as infinite and the above equation becomes -

$$F(I,t) = F(\infty,t) - F(\infty,I+t)$$

This shows that the integral decay heat per unit fission rate at a time t following an irradiation of duration I may be evaluated from the difference in the decay heat at cooling times of t and I+t following an infinite irradiation.

Because of these properties the infinite irradiation has become a valuable tool in decay heat evaluation and is capable of providing decay heat estimates for a wide range of irradiation and cooling times.

3.0 METHODS OF CALCULATION

Until about 1970 the bulk of the decay heat estimates used in the nuclear industry were obtained from the decay heat standards which were based upon the infinite irradiation decay heat functions described above. However, these standards related to U235 only. Since generally the U235 decay heat, at a given cooling time, is greater than that from either Pu239 or Pu241 the use of these standards yielded pessimistic decay heat estimates. Also, the standards failed to take account of neutron capture effects in fission products which can give rise to a significant increase in the decay heat levels at cooling times of a few years or more. In previous years the majority of applications required pessimistic estimates of the decay heat as provided by the decay heat standards. However, in some applications there has been a move recently towards best estimate calculations which require corresponding decay heat best estimates. It has therefore become necessary to consider alternative methods of calculating decay heat.

With the development of high resolution gamma radiation detectors and on-line mass separators in the early 1970s much data were obtained on the radioactive decay of short lived fission products. In parallel with this the power and speed of computers improved dramatically and the industry saw the development of summation codes for the prediction of decay heat. These codes simulate the irradiation conditions of reactor fuel, taking account of the fission distribution in U235, U238, Pu239, Pu241 etc., in order to predict the inventory of all fission products. An estimate of the decay heat is then obtained by summing over the beta and gamma energy release of all the nuclides present.

There are many codes available for carrying out such calculations. In the UK the main ones are FISPIN (Burstall, 1982), RICE (Nair,1977) and FISP6 (Tobias, 1982). The first two of these take account of the decay heat from both fission products and heavy elements while the third considers fission products only. Different methods of solution are also employed by these codes. FISPIN and RICE use numerical integration while FISP6 retains an analytical solution with linearised decay chains. For details of the mathematical equations which describe the build-up and decay of both the fission products and heavy elements the appropriate references should be consulted.

Decay heat predictions obtained with summation codes should be independent of the mathematical techniques employed. Discrepancies in results from two codes using nominally identical data libraries can be traced, with few exceptions, to differences in the basic data. Small differences are bound to occur between one computer installation and another as a consequence of the storage precision and rounding errors of each machine. However, it is widely accepted that these will be small and that decay heat predictions will be solely dependent upon the data libraries used. The constituent items of these data libraries will now be examined with particular reference to the yield and decay data available in the JEF1 library.

4.0 BASIC DATA REQUIREMENTS

The principal data requirements for fission product decay heat predictions fall into two major groups - fission yields and decay data. Because of the large neutron excess in the fissioning nuclide the fission products produced are neutron rich, often far from the line of stability, and decay via beta emission (and isomeric transitions) to a stable or extremely long lived nuclide. A typical decay chain, for atomic mass number 137, is shown in Figure 1.

4.1 Fission Yields

For many years in the UK there has been a continuing fission yield evaluation in progress, first by E. Crouch at Harwell and more recently by M. James and J. Banai (University of Birmingham) at Winfrith. This evaluation provides, for different fissile species, a best estimate of the independent fission yields for all known fission products and is based upon an extensive data base which has been built up over many years. In summarising the steps to produce a fission yield evaluation particular reference is made to the work of Banai and James (1986).

The construction of the data base requires great care and patience. All available fission yield measurements must be accurately recorded and a careful assessment made of their uncertainties. To a large extent this latter process has relied upon the judgement of the evaluator based upon his experience. It is also necessary to ensure that reported measurements are not duplicated in the data base since frequently the same results are quoted in a number of progress reports.

The first stage in deriving independent yields is to obtain best estimates of the chain yields (independent yields summed over each mass

chain). These correspond to the weighted mean value of all the reported yields for each mass chain after various corrections for radioactive decay, reference yields, etc. (Banai and James, 1986). For some fissile nuclides there are gaps in the data where no yield measurements have been reported and, in such cases, it is necessary for the evaluators to estimate the 'missing' chain yields by suitable interpolation or extrapolation.

The next stage is to derive independent fission yields. Because of the the lack of measured data this is achieved by applying a semi-empirical model of the independent yield distribution to the best estimate chain yields.

It has been well established that the independent yields within a mass chain are approximately fitted by a Gaussian charge dispersion -

$$P(A,Z) = \frac{1}{\sqrt{2\pi\sigma^2}} \exp\left\{-1/2\left(\left[Z - Zp(A)\right]/\sigma\right)^2\right\}$$

where

A is the mass number,
Z is the atomic number,
P(A,Z) is the probability of formation of nuclide (A,Z),
Zp(A) is the most probable nuclear charge in mass A
and σ is the charge dispersion parameter.

The fractional independent yield for nuclide (A,Z) is then

$$FIY(A,Z) = \int_{Z-1/2}^{Z+1/2} P(A,Z)\, dZ$$

Both Zp(A) and σ are functions of the mass number A. Where sufficient experimental data exist their values can be obtained from a probability plot of cumulative yield as a function of A. In many cases there are insufficient data to permit values of Zp(A) and σ to be obtained and some form of extrapolation or interpolation is required. Additional refinements to the charge dispersion model can take account of Z-even and Z-odd effects which have been observed experimentally.

The final process is one of adjustment in which the derived independent fission yields are constrained to satisfy a number of physical laws, eg.

- total number of fission fragments per fission = 2
- conservation of nucleons
- yields of complementary elements are equal
- conservation of nuclear charge
- fractional independent yields for each A sum to unity.

The procedures summarised above have been used by Banai and James (1986) to produce UKIFYA1 - the most recent UK fission yield evaluation for decay heat applications.

The JEF1 independent fission yield data library is based upon UKIFYA1 and covers all fission products with atomic numbers 25 to 70 and mass numbers in the range 72 to 175; it content is summarised in Table 2. Because the average neutron energies for current designs of fast reactors are much lower than those used in the bulk of the fast fission yield measurements it is widely considered that thermal fission yields are more representative for fast reactor calculations than are fast yields.

4.2 Decay Data

For the calculation of decay heat the parameters required for each radioactive fission product are its half-life and estimates of the average energy release per disintegration in the form of 'beta' and 'gamma' radiation. The 'beta' component should include contributions from conversion and Auger electrons as well as beta particles while the 'gamma' component should include any X-ray contributions. Also, when a nuclide decays to two or more daughter products the appropriate branching fractions are required.

Radioactive half-lives and many branching fractions are determined directly from measurements. However, the same is not true of the average decay energies required for decay heat calculations. The result of most radioactive decay studies is a decay scheme which describes the relative emission probabilities of gamma transitions that arise in the de-excitation of the daughter nucleus. Occasionally, details of the beta spectrum have also been studied experimentally and the relative branching of the beta transitions to the excited levels of the daughter nucleus determined. In most cases however the relative beta branching has been inferred from the measured gamma data. It must be emphasised that, even in many of these cases, the decay scheme data are incomplete from the point of view of decay heat calculations and it is the task of the evaluator to derive the required parameters by suitable deduction.

The average gamma energy release per disintegration may be obtained by summing the products of the gamma energies with their emission probabilities. However, the absolute emission probabilities will not always be known or may be highly uncertain. To this must be added the average energy released in the form of X-rays per disintegration.

There have been relatively few measurements made of the distribution of conversion electrons, Auger electrons and X-rays following the internal conversion of gamma radiation or following electron capture decay. Tabulations of predicted internal conversion coefficients (eg. Hager and Seltzer, 1968) as a function of energy for each atomic number are available for gamma transitions of different multipolarities. Thus, the distribution of X-rays and electrons arising from internal conversion may be calculated. For the atomic K-shell the calculations are relatively straightforward but become more complex for the L- and higher electron shells.

Most measurements of beta decay yield estimates of the beta end point energies and corresponding emission probabilities. What is required for decay heat calculations is the average energy emitted in the form of beta radiation per disintegration. Strictly, this should be derived from

the observed distribution of beta particles as a function of energy. Since these data are rarely available it is necessary for the evaluator to calculate the average beta energy of each transition from beta decay theory. This approach is valid for allowed and unique forbidden beta transitions but for non-unique transitions measurements must be made to provide the correct data.

In the absence of such data there is a popular practice of calculating average beta energies of such transitions by treating them as unique transitions of a lower degree, ie. a first forbidden non-unique transition would be treated as allowed and a second forbidden non-unique transition would be treated as first forbidden unique. However, it must be emphasised that the validity of this approach has yet to confirmed.

These comments illustrate that even for relatively simple and well defined decay schemes the evaluators need to provide considerable effort to obtain the required parameters for decay heat calculations. Complex decay schemes, as are found for many fission products, give rise to additional problems.

The absolute normalisation of gamma emission probabilities depend upon the magnitude of the beta branch to the ground state of the daughter nucleus. There are many instances where this has not been accurately determined. Also, many experimental studies have concentrated on the most intense radiations at the lower energies. It has been argued that, as a result, in the study of complex decay schemes with large Q-values the beta branching to highly excited states may be 'missed' (Hardy et al, 1977). This is because, invariably, the beta branches are deduced from the difference in intensities of gamma rays populating and depopulating the various excited levels. In the construction of the decay scheme certain gamma rays are unplaced while others may remain undetected because of their high energy or relatively low intensity. As a consequence the average beta energy is overestimated with a corresponding underestimate of both the average gamma energy and the combined beta+gamma energy.

Various approaches have been adopted in order to overcome the problems with such nuclides. For example, Yoshida (1982) has used the gross theory of beta decay to predict the average decay energies of these fission products and extended it to include a number of nuclides with decay schemes which are believed to be well defined. Aleklett and Rudstam (1982) have measured directly the average beta energies of some fission products with 'well known' decay schemes and have used measured beta strength functions, or extrapolations to them, to predict the corresponding average gamma energies. These beta strength functions have also been used by Aleklett and Rudstam (1982) and Reich and Bunting (1982) to obtain estimates of the average decay energies of short-lived fission products for which little decay data have been measured. Microscopic calculations of the beta strength function have been made by Klapdor (1983, 1985) and corresponding estimates of the average decay energies obtained. Mann et al (1982) have demonstrated the use of a statistical model for the prediction beta decay properties. The present paper makes no attempt to examine the virtues or disadvantages of these methods but merely notes the diversity of methods which may be applied.

The JEF1 decay data file covers a wide range of nuclides which includes the fission products. Data for this were selected from existing UK and French evaluations on the basis of a consistency parameter derived for each nuclide. This was taken as the percentage deviation between the Q-value of the decay and the sum of the component radiation emissions and gives a measure of the consistency of the derived decay scheme. Where data for any nuclide were available from both evaluations the one which gave the most consistent decay scheme was selected for JEF1. Table 3 summarises the content of the entire JEF1 decay data file in terms of the numbers of nuclides taken from the two evaluation sources. As noted in the above discussion, it is not always possible to estimate the average decay energies of short lived fission products from the derived decay scheme data. In such cases, estimates of the average decay energies were taken from the work of Aleklett and Rudstam (1982) or from Yoshida (1983) when these were not available.

Table 4 summarises the fission product decay data included in JEF1 and compares the general statistics with those of UKFPDD-2. The smaller number of radioactive nuclides in JEF1 is due to the exclusion of some short lived fission products which were included in UKFPDD-2 and whose decay parameters were all estimated theoretically. These nuclides are listed in Table 5. It should also be noted that there are significantly more nuclides with spectral data in JEF1 compared to UKFPDD-2.

4.3 Cross Sections

In addition to the fission yield and decay data summarised above the calculation of decay heat also requires neutron capture cross section data for the fission products. There are a number of nuclides which have very small direct fission yields and whose primary production route is through neutron capture in stable isotopes of the preceding mass chain. Important nuclides in this category include Cs134, Pm148, Pm148m and Eu154. The form of the cross section data used by the various summation codes differs. Some use the 2200 m/s thermal cross sections and resonance integrals, as are available in JEF1, while others use spectrum averaged cross sections appropriate to the reactor system being studied. For the present purposes it is sufficient to note that these basic data are available in JEF1.

Differential fission and capture cross section data for the Actinides are also required for decay heat calculations but their use is indirect. These data are processed to a multi-group structure for use by the reactor physics lattice codes in the calculation of neutron spectra and burn-up dependent relative fission rates in the various fissile species present in the fuel - U235, U238, Pu239 and Pu241. The summation codes utilise these results, in a variety of different forms, for calculations relating to specific reactor types. These data are also used in calculating the magnitude of the heavy element decay heat contribution. For cooling times of less than approximately 10 days this arises principally from the radioactive decay of U239 and Np239. At much longer cooling times (greater than a few years) the decay of higher Actinides become important.

5.0 DECAY HEAT CALCULATIONS WITH JEF1

In examining the results of decay heat predictions using JEF1 it will be of interest to examine the effect of both the fission yields and decay data separately. For the purpose of comparisons, use will be made of the decay heat predictions obtained using the currently recommended UK decay data file UKFPDD-2 (Tobias and Davies, 1980) and fission yield data Crouch 3I (Crouch, 1977, 1980). All decay heat predictions described in this note were obtained with the summation code FISP6 (Tobias, 1982).

5.1 The effect of 'theoretical' nuclides

It was noted earlier that no provision was made for including in JEF1 a number of short-lived 'unknown' fission products for which all decay parameters had been theoretically estimated. These nuclides (listed in Table 5) are included in UKFPDD-2 (from US-ENDF/B-IV data sources) but are absent from JEF1. In order to examine the effect on decay heat predictions of these nuclides the UKFPDD-2/Crouch 3I data library for FISP6 was modified so that all theoretical nuclides were assigned zero yields and that the first 'known' nuclide of each mass chain was assigned the appropriate cumulative yield. Figure 2 shows the changes in decay heat predictions obtained by excluding these theoretical nuclides for fission pulses in U235 and Pu239 respectively. The differences observed are generally small (a few percent or less, and mainly at very short cooling times) indicating that the exclusion of short-lived theoretical nuclides from JEF1 will have a negligible effect on integral decay heat predictions beyond 20 s cooling. For cooling times of less than 20 s differences of only 1 or 2 % in integral decay heat estimates will occur.

5.2 The effect of JEF1 Fission Yields

The effect on decay heat predictions of a change in fission yield data only was examined. For this purpose comparisons were made between predictions obtained using UKFPDD-2 decay data with both the Crouch 3I and JEF1 fission yields. Figures 3 and 4 show the decay heat ratios with JEF1/Crouch fission yields for predicted fission pulses in U235 and Pu239. The greatest differences are seen generally for Pu239. At short cooling times these are due to changes in a large number of independent fission yields while at longer cooling times they are due to changes in a relatively small number of chain yields. Differences of up to a few percent in Pu239 chain yields are found for mass chains 90, 137, 103, 106 and 140. Since the JEF1 fission yields are based upon an extension of the Crouch 3I data base, using a more rigorous mathematical treatment than was used previously (Banai and James, 1986),they are the preferred data.

5.3 The use of JEF1 decay data and fission yields

Figures 5 and 6 compare the results of JEF1 and UKFPDD-2/Crouch 3I decay heat predictions for the fission pulse and infinite irradiation in both U235 and Pu239. For the fission pulse the greatest differences are found at short cooling times (less than 100 s) while at longer times the differences are similar in magnitude to those due to the fission yield

data. The differences in decay heat predictions for the infinite irradiation cases are seen to be much smaller, being typically 0.5-1 %. These comparisons show that the use of JEF1 decay and fission yield data, in preference to UKFPDD-2 with Crouch 3I fission yields, will result in slightly reduced (by about 0.5 to 1 percent) integral decay heat estimates.

It was noted above that a significant number of the short lived fission products in JEF1 have been assigned average decay energies which are not based upon decay scheme data. As a consequence, the predicted gamma decay heat at short cooling times will not be totally accounted for by the corresponding predicted gamma spectrum. Figures 7 and 8 show this difference as a function of cooling time for fission pulses and infinite irradiation respectively in U235, U238, Pu239 and Pu241. In both figures it is seen that by 1000 s cooling the contribution to the gamma decay heat from the these short lived nuclides has become insignificant. The corresponding beta decay heat components are expected to display similar characteristics. It should be noted that these results show considerable improvement over those for UKFPDD-2 with Crouch 3I yields (Tobias and Davies, 1980).

As a consequence of the results given in Figures 7 and 8 it is not unreasonable to expect that, in comparisons with measured decay heat data, the largest discrepancies will be found at the shorter cooling times where this group of nuclides make a significant contribution to the predicted levels.

6.0 COMPARISONS WITH DECAY HEAT MEASUREMENTS

The performance of decay heat predictions may be gauged through comparisons with measured data. In recent years there have been a number of 'benchmark' measurements made for U235 and Pu239 decay heat; these are summarised in Tables 6 and 7 respectively. It will be seen from these tables that the measurements have been made for a wide range of irradiation and cooling times (with varying precision). Comparisons between these individual measurements and corresponding predictions using UK data files have been examined in detail by James (1983).

The properties of the fission pulse function described earlier show that the various decay heat measurements correspond to integrals of the appropriate pulse function evaluated over limits defined by the irradiation and cooling times. It follows that by 'unfolding' the various decay heat measurements they can all be converted to the same basis - the fission pulse function, each one covering a range of cooling times defined by those of the measurements.

The basis of such a method was described by Schmittroth and Schenter (1979) in which the fission pulse function is represented by -

$$f(t) = \sum_i x(i) \exp(-\lambda_i t)$$

Since the exponential terms can be easily integrated it is possible to apply this to a variety of decay heat measurements for different

irradiation times. By minimising the sum of squared residuals (the difference between the measurement and the model f(t)) the best estimate of the least squares parameters x(i) can be obtained. Thus, application of this technique to each set of decay heat measurements will yield a set of parameters x(i) that will describe the portion of the fission pulse function it represents. The range of cooling times for which each unfolded pulse function is valid is defined by the shortest cooling time for which the measurements were made and the sum of the longest cooling time plus the irradiation time. By applying a least squares analysis to these unfolded pulse functions, and taking account of correlated uncertainties, a best fit to the available measurements has been obtained for both U235 and Pu239 (Tobias, 1986). These derived 'benchmarks' greatly ease the analysis of decay heat predictions since they eliminate the need for individual comparisons for each set of measurements with its own irradiation conditions and cooling times.

Figure 9 compares both JEF1 and UKFPDD-2/Crouch 3I beta+gamma decay heat predictions for a fission pulse in U235 with the corresponding best fit data derived by Tobias (1986). There is seen to be a marginal improvement in prediction, relative to the best fit data, at short cooling times through the use of the JEF1 data. As expected, the largest differences are seen to occur at cooling times of less than 100 s. However, the discrepancy at 1000-2000 s is unexpected since, at these cooling times, the principal decay heat nuclides have decay schemes which are believed to be reasonably well establised.

Similar comparisons for the Pu239 decay heat pulse function are given in Figure 10. Again, there appears to be a marginal improvement, relative to the best fit data, with the JEF1 data. The discrepancy between prediction and best fit at short cooling times appears to be smaller than for U235. However, at cooling times of 200 - 2000 s it is greater.

It should be noted that the best fit data for U235 and Pu239, shown in Figures 9 and 10 respectively, were obtained from a least squares analysis of measured data only and were not normalised via the corresponding predictions. Consequently, the good agreement found between these data and the predictions at the longer cooling times is highly encouraging.

Figure 11 compares the pulse function predictions for U238 with corresponding values derived from the recent measurements of Akiyama et al (1985, 1986). A striking feature of these comparisons is again the discrepancy between measurement and prediction at cooling times of 200-2000 s. On the whole there appears to be relatively little difference between the JEF1 and UKFPDD-2/Crouch 3I predictions relative to the measured data except perhaps at cooling times of around 10,000 s.

Comparisons between Pu241 pulse function predictions and corresponding values from the measurements of Dickens et al (1981) are given in Figure 12. The discrepancy between measurement and prediction at 1000 s cooling is again evident while the differences between the two sets of predictions are similar to those found for U238.

The comparisons of Figures 9-12 show that, in general, the differences in decay heat predictions for JEF1 and UDFPDD-2/Crouch 3I are relatively small and that there is little to choose between them. In all cases there appears to be a discrepancy between measurement and prediction at cooling times of approximately 1000 s. Since, at this cooling time, the principal nuclides with respect to total decay heat have decay schemes which are believed to be well established the comparisons suggest that some may be in error. The possibility of the discrepancy being due to errors in chain yields is less likely since changes of up 50 % would be required.

Table 8 lists the principal decay heat contributors at a cooling time of 1000 s following fission pulses in U235, U238, Pu239 and Pu241 and includes details of their average decay energies and Q value from JEF1. In each case about 75% of the predicted decay heat is accounted for by the 19 nuclides listed. Although the same nuclides appear in all 4 lists their relative contributions vary considerably. It has been previously noted that evaluations for a number of the nuclides listed in Table 8 rely on single or relatively few sets of measurements (Dickens, 1983). In many cases the measurements are over 10 years old.

From the earlier discussion on problems in decay scheme measurements it is apparent that the nuclides most likely to be in error will have overpredicted average beta energies and correspondingly underpredicted average gamma energies. If the decay data for a nuclide was in error to the extent that the average gamma energy was underestimated by 1.0 MeV then, because of the energy removed by the anti-neutrino, the average beta energy would be overestimated by a fraction of this, say 0.4 MeV. Thus, correction of this decay scheme would result in an increase of 0.6 MeV in the average beta+gamma energy. It should be noted that these arguments are consistent with the observed discrepancies between measurement and prediction for the separate decay heat components (Tobias, 1983; James, 1983). ie. at cooling times of approximately 1000 s there is a tendency to underpredict the gamma component with a corresponding, but less pronounced, overprediction of the beta component.

From Table 8 it is apparent that nuclides such as Mo101 and I134, which have relatively large average gamma energies compared to the Q values, have less scope for significant changes (should their decay schemes be subject to the errors noted above) than other nuclides such as Tc102 and Xe137. Also, the decay schemes of a number of the nuclides in Table 8 have substantial ground state beta branches so that any change in their magnitude could result in significant revision of their average beta and gamma decay energies. Further experimental studies of these nuclides are required in order to resolve the observed discrepancy.

It should be noted that this discrepancy could also arise as a consequence of incorrect half life assignments for other nuclides. However, the magnitude of such errors is such that this possibility is considered to be less likely than that suggested above.

The comparisons described in this note show that general improvements in decay heat predictions for direct fission products may be achieved by

further decay scheme studies on short lived nuclides (half life < 100 s). There is also evidence to suggest that a number of nuclides with half-lives of around 1000 s have been poorly characterised in terms of their decay properties and that further experimental studies are needed for some of them.

7.0 CONCLUSIONS

The data requirements, in terms of decay data and fission yields, for decay heat calculations have been examined and the evaluation procedures used are described. The data available in the recent JEF1 files are summarised and their use in decay heat calculations has been studied. From comparisons of JEF1 decay heat predictions with both UK data predictions and measured data it was found that :-

1. The exclusion from JEF1 of short lived 'theoretical' nuclides , previously in the UK decay data library, has only a relatively small effect on decay heat predictions at short cooling times following fission pulses. The corresponding effect on integral decay heat predictions is negligible.

2. A change in fission yield data, from UK to JEF1, results in differences of up to a few percent in decay heat predictions for a wide range of cooling times following fission pulses. The effect on integral decay heat predictions is much smaller. The differences were attributed not only to changes in independent fission yields (at short cooling times) but also to those in mass chain yields (at longer cooling times). The JEF1 fission yields are the preferred data in view of their more recent data base and more rigorous mathematical derivation.

3. The use of JEF1 decay and fission yield data resulted in additional small differences in fission pulse predictions at short cooling times. On the whole, differences in prediction with JEF1 and UK data could be attributed largely to the change in fission yield data. Changes in decay data had a less marked effect on decay heat predictions although it was noted that JEF1 decay heat predictions are more complete than those with current UK data files in terms of the gamma spectra calculated at short cooling times.

4. Discrepancies at cooling times of less than 100 s were noted between JEF1 (and UK) fission pulse predictions and corresponding values derived from measurements. These can be attributed to the acknowledged deficiencies in decay data for short lived fission products. Underpredictions of up to 10 % in total decay heat were also observed at a cooling time of 1000 s following a fission pulse. The magnitude of these discrepancies is such that they are more likely to be due to errors in decay data for a small number of nuclides than to errors in fission yields. The principal nuclides contributing to the decay heat at this time were identified as Rb89, Sr93, Y94, Y95, Mo101, Tc102, Tc104, Tc105, Sb130, Sb131, Te133, I134, Xe137, Xe138, Cs138, Cs139, Ba141, Ba142 and La143. The effect of these discrepancies on corresponding integral decay heat predictions are much smaller but, in certain applications, may lead to uncertainties which exceed the required accuracies.

It was noted that the decay data for a number of these nuclides had been taken from single or relatively few measurements and also that a number of them had average gamma energies that were relatively small compared to their Q values. Recognised problems in decay scheme measurements suggest that this group of nuclides offers the greatest scope for revision should their decay schemes be in error. Further experimental studies of these nuclides are required in order to resolve the observed discrepancy.

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Table 1. Accuracy Requirements for Decay Heat Predictions

System	Required Accuracy %					
	0	1 min	10 min	8 hr	24 hr	days	months
PWR - BWR	<---------- (5) ---------->					
Gas-Cooled	<---------- (5) ---------->					
Fast	<------- (integrated 10) --------->					
Fuel Handling/Storage				<-------- (5) -------->	<-- (<5) -->	

Table 2. Summary of JEF1 Adjusted Independent Fission Yields

Fissile Nuclide	No. of fission products		
	Thermal	Fast	14.5 MeV
Th232		745	758
U233	710	706	770
U235	768	748	744
U238		740	815
Pu239	769	757	
Pu240		761	
Pu241	751	752	

Table 3. Summary of JEF1 Decay Data File

Total no. of nuclides	=	1128
Ground state	=	907
1st excited state	=	210
2nd excited state	=	11
Nuclides with spectral data	=	1021
Data from UK evaluations	=	419
Data from French evaluations	=	709
Total no. of gamma lines	=	23157
Total no. of beta- lines	=	5769
Total no. of beta+ lines	=	651
Total no. of alpha lines	=	1394
Total no. of electron electrons	=	10869
Total no. of X-rays	=	2345

Table 4. Summary of JEF1 Fission Product Decay Data

Number of radioactive nuclides	=	585 (736)
Ground state	=	444 (596)
1st excited state	=	136 (133)
2nd excited state	=	5 (7)
Nuclides with spectral data	=	450 (390)

Note: UKFPDD-2 values given in parenthesis

Table 5. Short-Lived Fission Products Omitted from JEF1 Decay Data

Symbol	Mass Nos.
Co	72 73 74 75
Ni	72 73 74 75 76 77 78
Cu	74 75 76 77 78 79 80 81
Zn	80 81 82 83
Ga	85
Ge	87 88
As	89 90
Se	93
Br	93 94 95 96
Kr	96 97 98
Rb	99 100 101
Sr	101 102 103 104
Y	103 104 105 106 107
Zr	105 106 107 108 109
Nb	107 108 109 110 111 112
Mo	109 111 112 113 114 115
Tc	111 113 114 115 116 117 118
Ru	114 115 116 117 118
Rh	115 116 117 118 119 120 121 122 123
Pd	121 122 123 124 125 126
Ag	124 126 127 128
Cd	127 129 130 131 132
In	133 134
Sn	-
Sb	139
Te	142
I	142 143 144 145
Xe	146 147
Cs	148 149 150
Ba	149 150 151 152
La	151 152 153 154 155
Ce	152 153 154 155 156 157
Pr	154 155 156 157 158 159
Nd	156 157 158 159 160 161
Pm	158 159 160 161 162
Sm	160 161 162 163 164 165
Eu	163 164 165
Gd	165

Table 6. Summary of Recent 235U Total Decay Heat Measurements

Expt.	Irrad.	Cooling Times	Reference
1 a	100 s	70 - 3000 s	CEA : Lott et al (1973)
b	1000 s	200 - 2.0E4 s	" "
c	5000 s	300 - 7.0E4 s	" "
2 a	2.0E4 s	10 - 1.0E5 s	LASL : Yarnell and Bendt (1977)
b	2.0E4 s	10 - 1.0E5 s	" " (1978)
3 a	4 h	600 - 1.0E4 s	UCB : Schrock et al (1979)
b	22 h	600 - 2.0E4 s	" "
4 a	1000 s	1 - 5.0E4 s	IRT : Friesenhahn and Lurie (1979)
b	2.0E4 s	1 - 1.0E5 s	" "
c	1 d	1 - 1.5E5 s	" "
d	35 d	1 - 2.0E5 s	" "
5 a	1 s	1.7 - 89.7 s	ORNL : Dickens et al (1980)
b	10 s	10.7 - 595 s	" "
c	100 s	70 - 9950 s	" "
6	200 s	15 - 4000 s	KFK : Baumung (1981)
7 a	10 s	11 - 3800 s	YAYOI: Akiyama et al (1982a, 1982b)
b	100 s	150 - 2.2E4 s	" "

Table 7. Summary of Recent 239Pu Total Decay Heat Measurements

Expt.	Irrad.	Cooling Times	Reference
1	Pulse	50 - 1.0E5 s	CEA : Fiche et al (1976)
2	2.0E4 s	20 - 1.0E5 s	LASL : Yarnell and Bendt (1978)
3 a	1000 s	1 - 5.0E4 s	IRT : Friesenhahn and Lurie (1979)
b	1 d	1 - 1.5E5 s	" "
4 a	1 s	1.7 - 100 s	ORNL : Dickens et al (1981)
b	5 s	17 - 800 s	" "
c	100 s	250 - 9950 s	" "
5 a	10 s	11 - 4000 s	YAYOI: Akiyama et al (1982a, 1982b)
b	100 s	950 - 2.0E4 s	" "

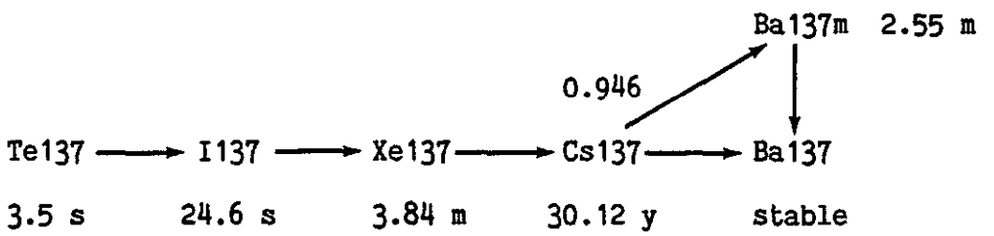
Table 8

Principal Decay Heat Nuclides at 1000 s Following a Fission Pulse

Nuclide		....% Decay Heat Contribution.... U235	U238	Pu239	Pu241	Q MeV	EB MeV	EG MeV
Rb89		6.96	4.42	2.45	1.72	4.486	1.007	2.068
Sr93		5.88	4.78	3.63	2.72	3.950	0.685	1.939
Y94	*	6.43	4.90	4.52	3.33	4.882	1.793	0.772
Y95	*	7.27	5.83	5.59	4.38	4.430	1.340	1.292
Mo101		4.20	3.26	5.30	4.79	2.811	0.526	1.473
Tc102	*	3.64	4.97	5.42	5.32	4.525	1.945	0.081
Tc104	*	2.51	10.2	8.58	9.05	5.400	1.578	1.728
Tc105	*	0.66	2.76	4.11	4.23	3.400	1.244	0.459
Sb130		2.58	1.68	2.88	3.44	4.970	0.977	2.491
Sb131		1.66	2.53	2.42	2.12	3.100	0.542	1.695
Te133	*	2.52	3.58	2.40	3.40	2.970	0.818	0.929
I134		1.72	1.04	2.51	1.74	4.150	0.622	2.541
Xe137	*	2.11	2.16	2.18	2.23	4.343	1.780	0.180
Xe138		4.87	4.47	3.87	4.38	2.830	0.671	1.126
Cs138		4.67	4.11	4.46	4.47	5.335	1.269	2.361
Cs139	*	5.57	5.12	5.01	5.75	4.290	1.681	0.308
Ba141	*	3.81	3.80	3.60	3.13	3.030	0.868	0.839
Ba142		3.43	2.82	2.86	2.85	2.200	0.426	1.045
La143	*	3.49	2.73	2.70	2.56	3.300	1.315	0.098
% Dev.		5.5	13.5	8.0	6.7			

* Nuclides for which average gamma energies are relatively small in comparison with Q value.

Figure 1. Mass 137 Fission Product Decay Chain



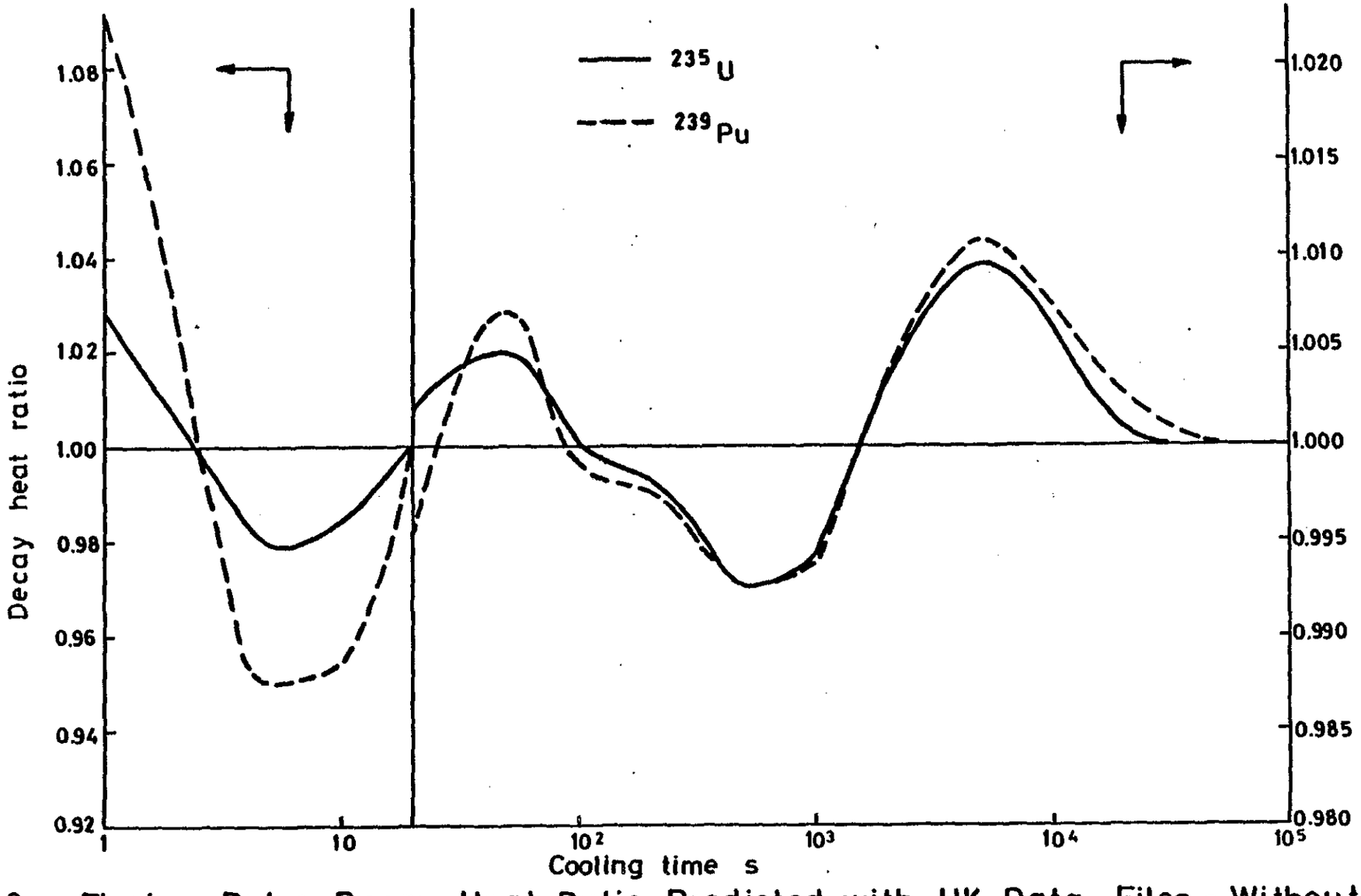


FIG. 2. Fission Pulse Decay Heat Ratio Predicted with UK Data Files - Without / With Theoretical Nuclides.

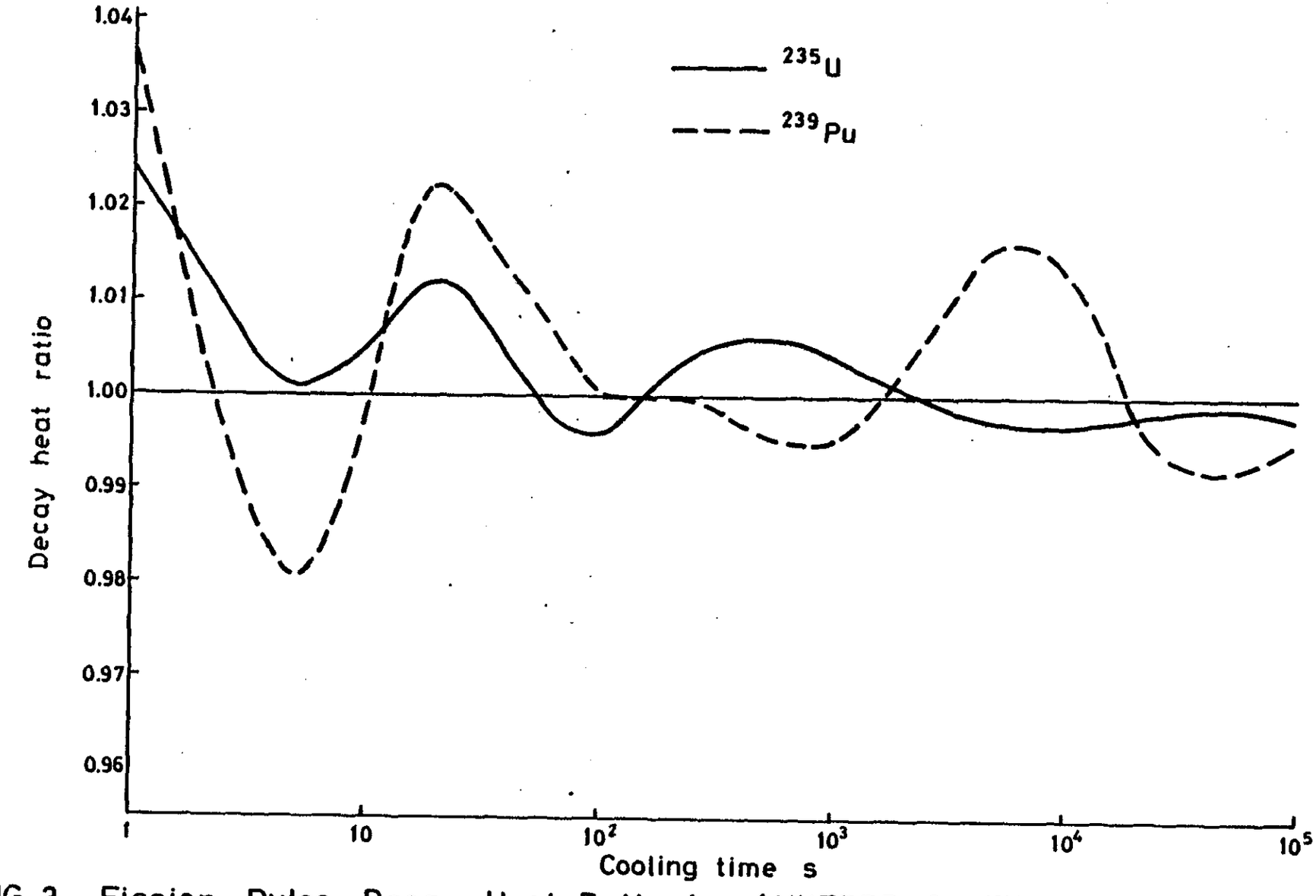


FIG. 3. Fission Pulse Decay Heat Ratio for (UKFPDD-2 + JEF1 Yields) / (UKFPDD-2 + Crouch 31 Yields)

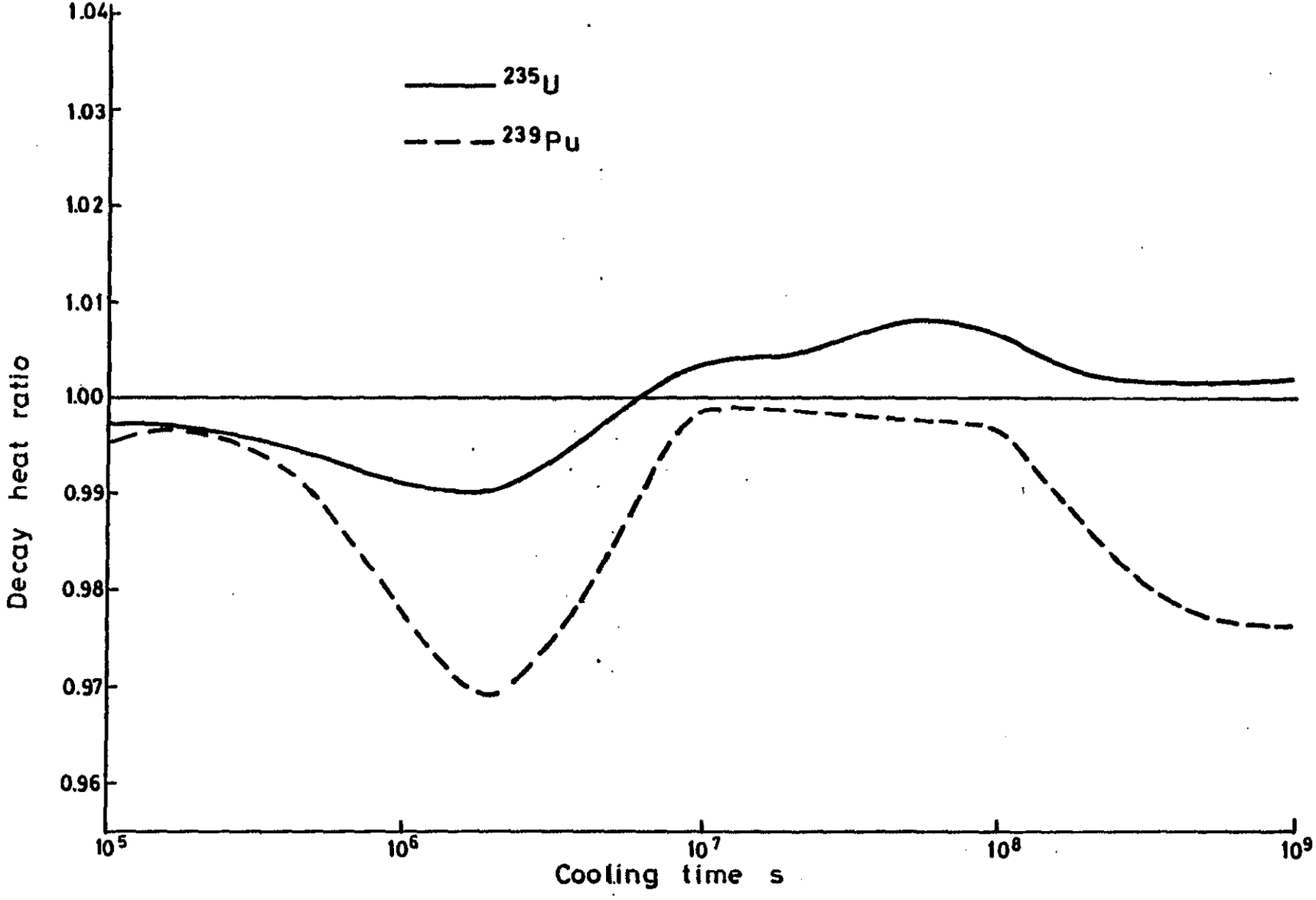


FIG.4. Fission Pulse Decay Heat Ratio for (UKFPDD-2 + JEF1 Yields) / (UKFPDD-2 + Crouch 31 Yields)

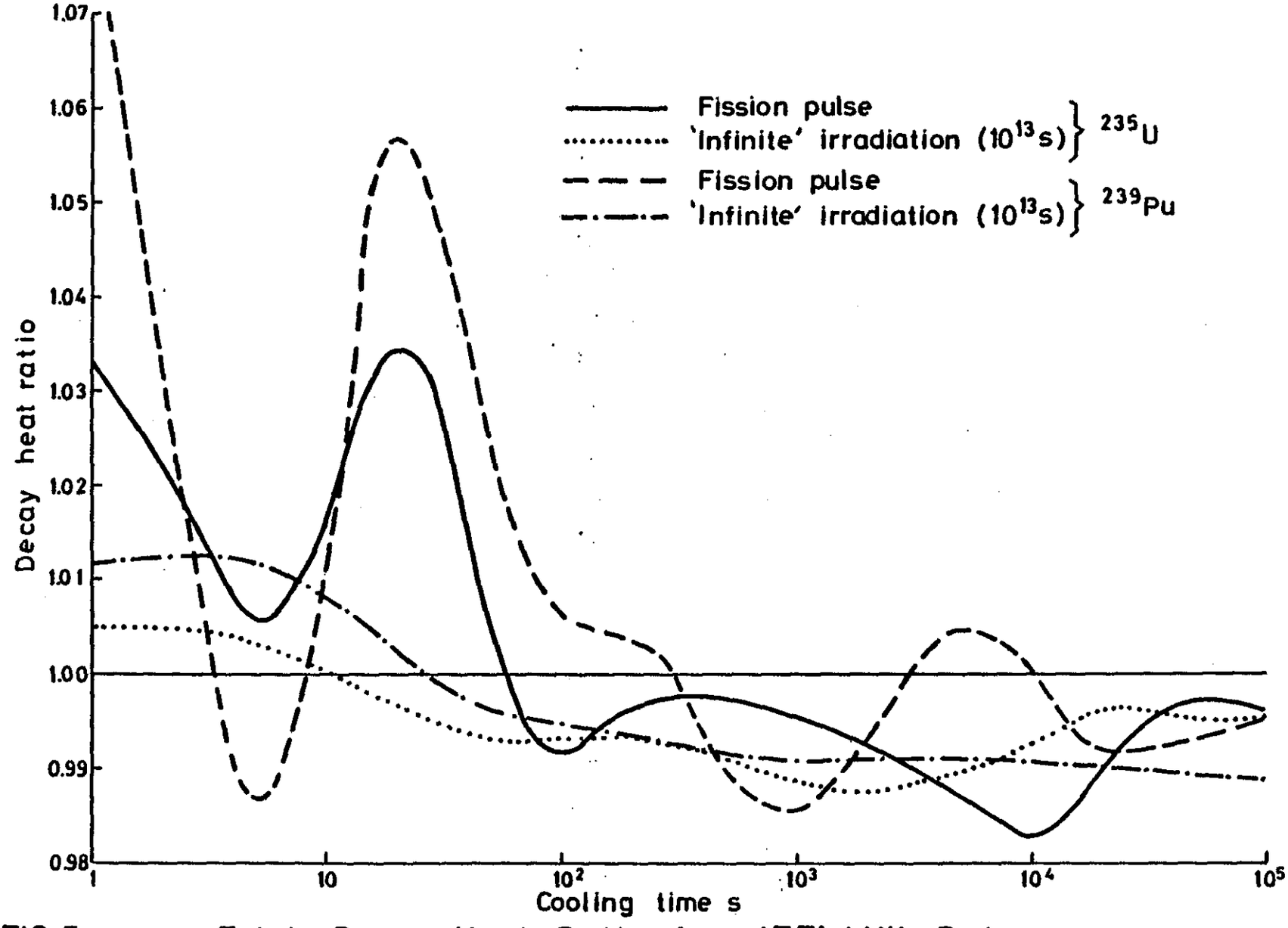


FIG.5. Total Decay Heat Ratio for JEF1 / UK Data.

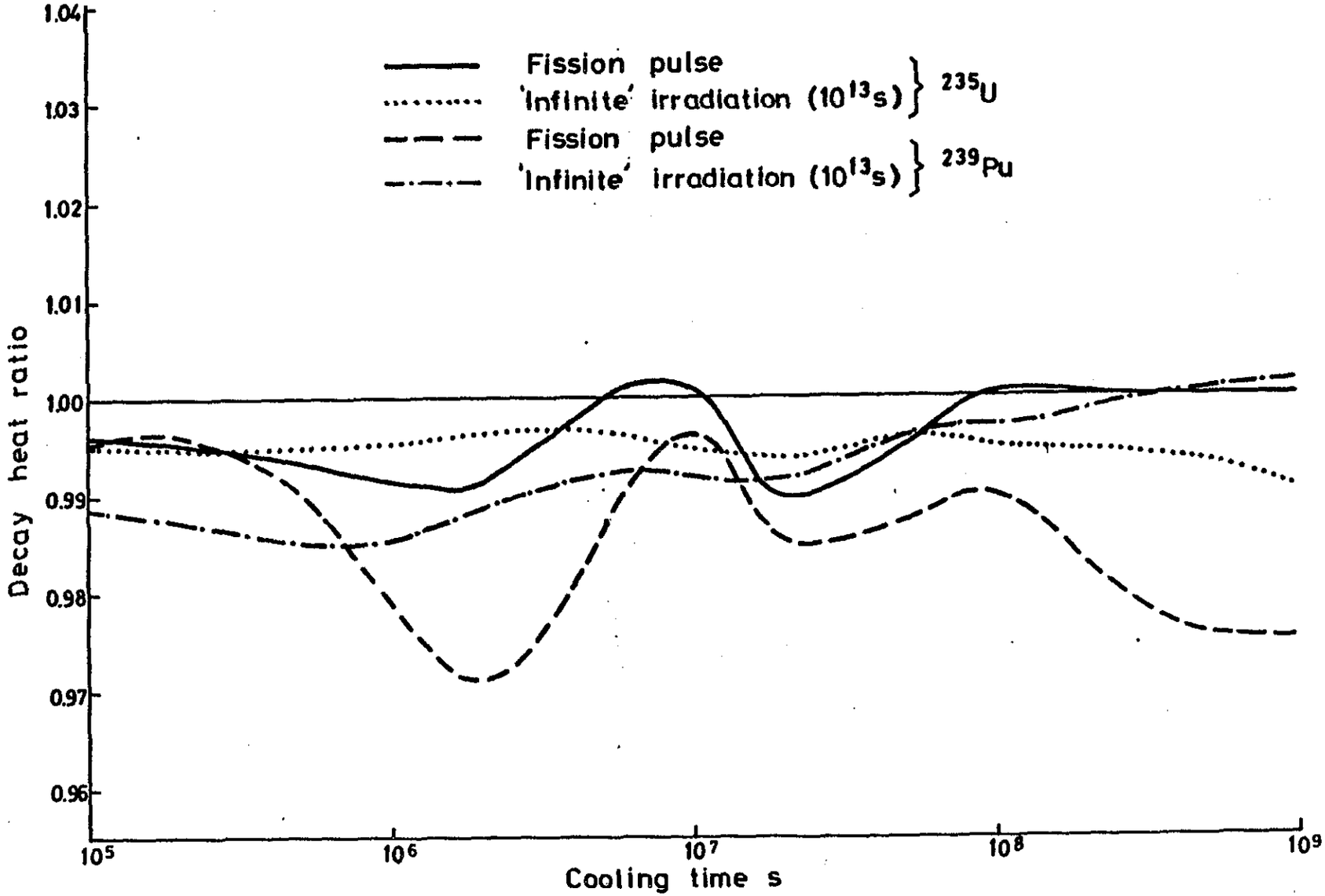


FIG. 6. Total Decay Heat Ratio for JEF1 / UK Data.

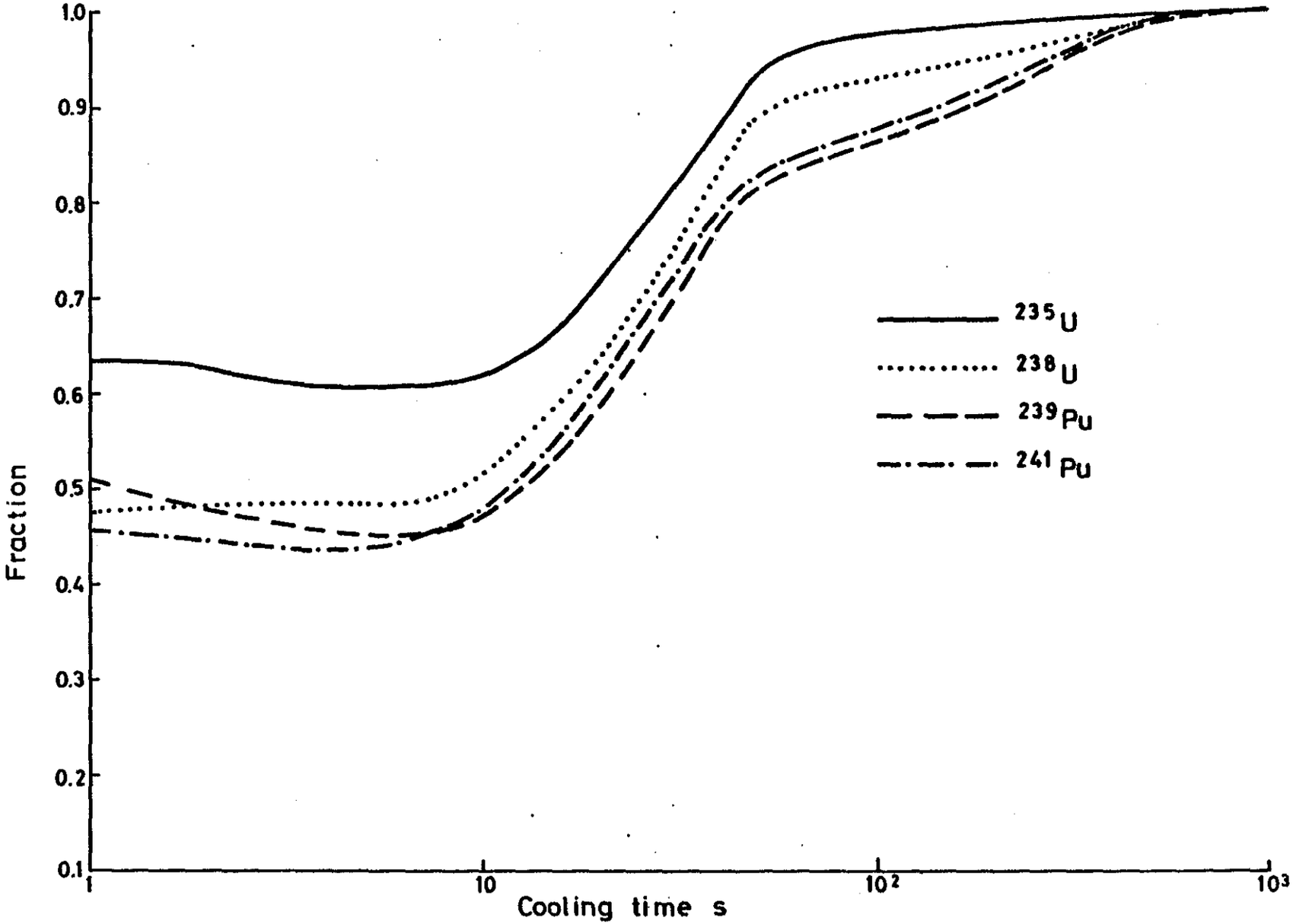


FIG.7. Fraction of Gamma Decay Heat Accounted for by Spectral Data in JEF1 Fission Pulse Predictions.

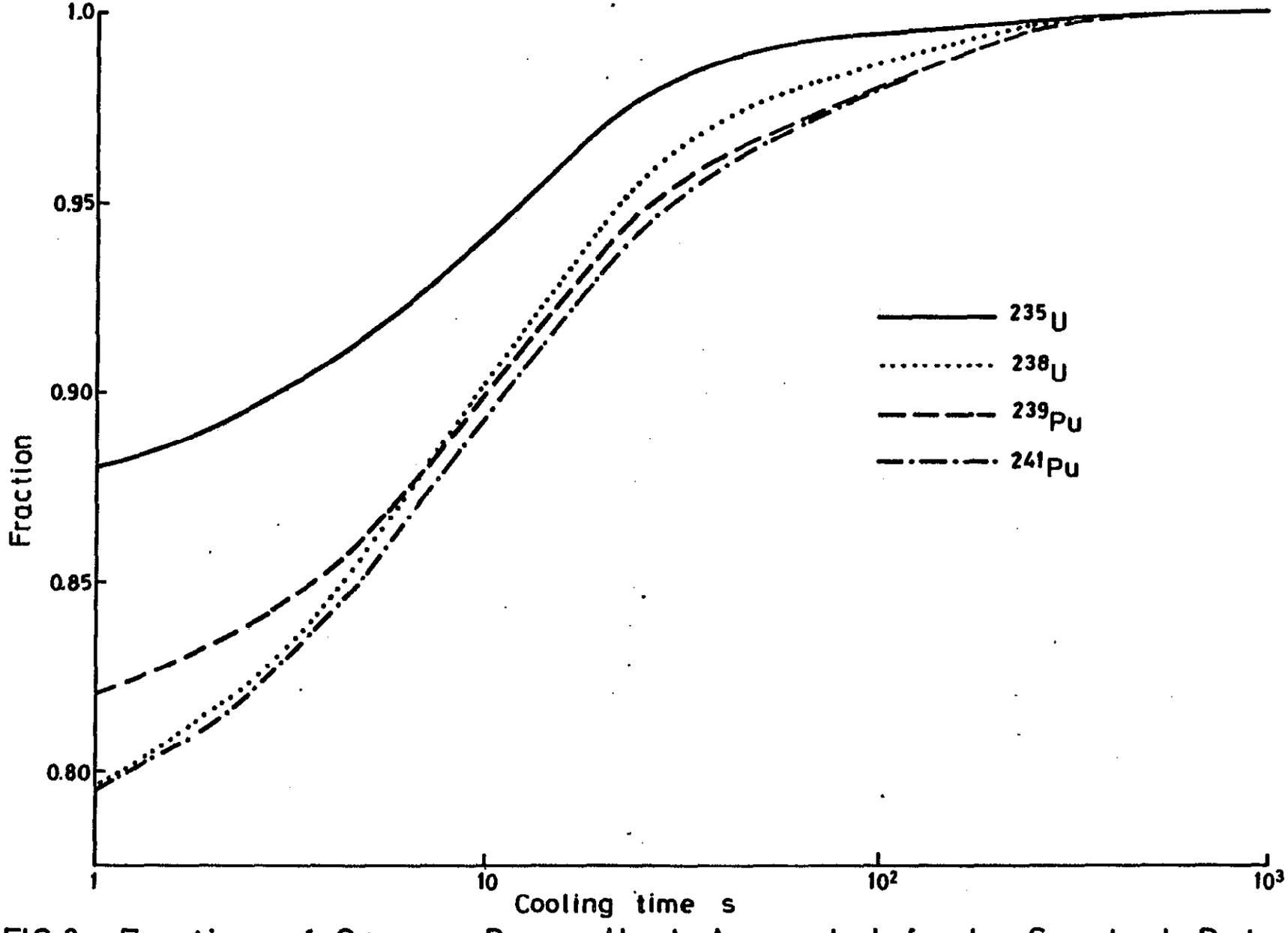


FIG.8. Fraction of Gamma Decay Heat Accounted for by Spectral Data in JEF1 Infinite Irradiation Predictions.

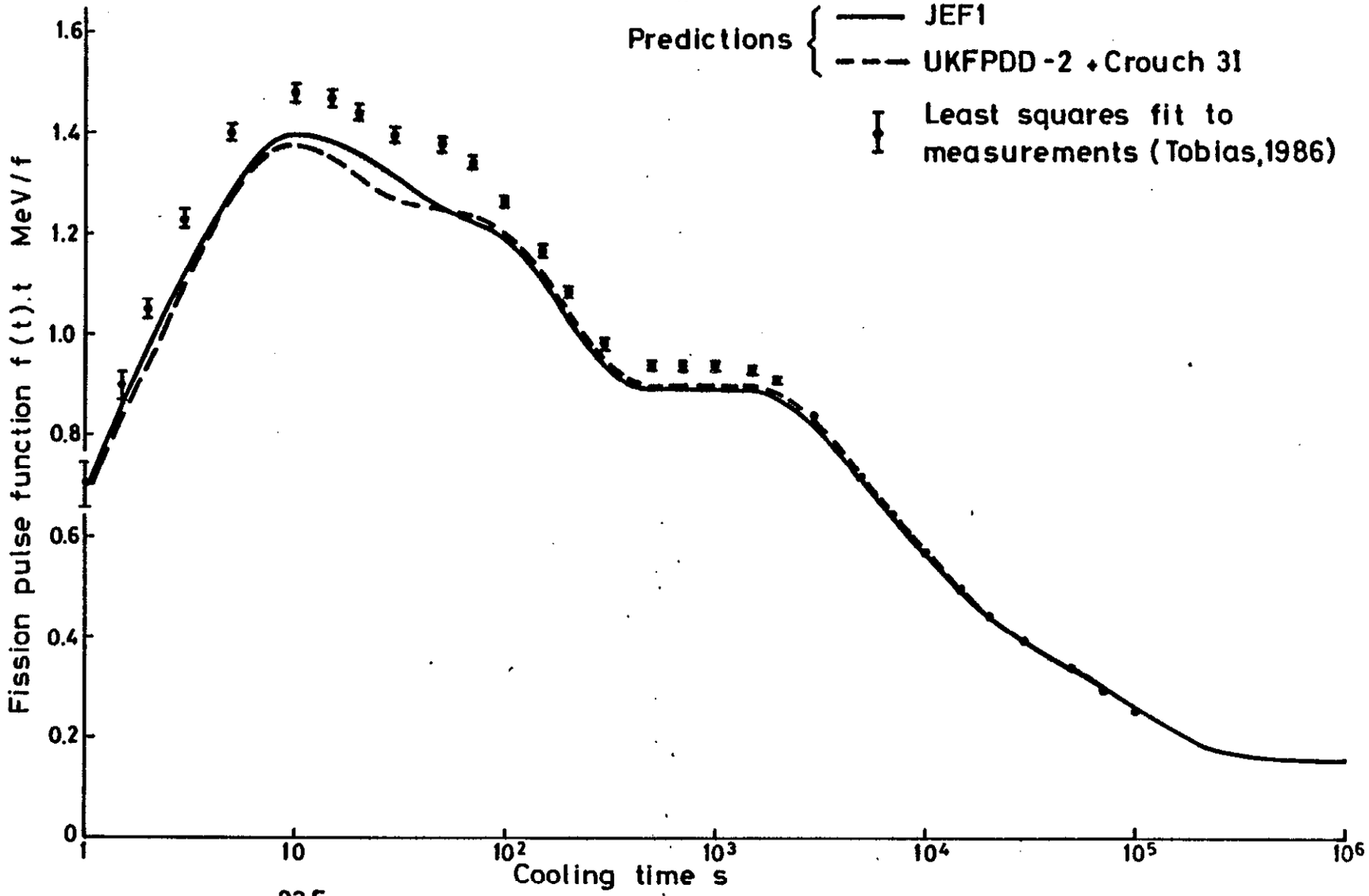


FIG. 9. ^{235}U Beta + Gamma Decay Heat - Fission Pulse.

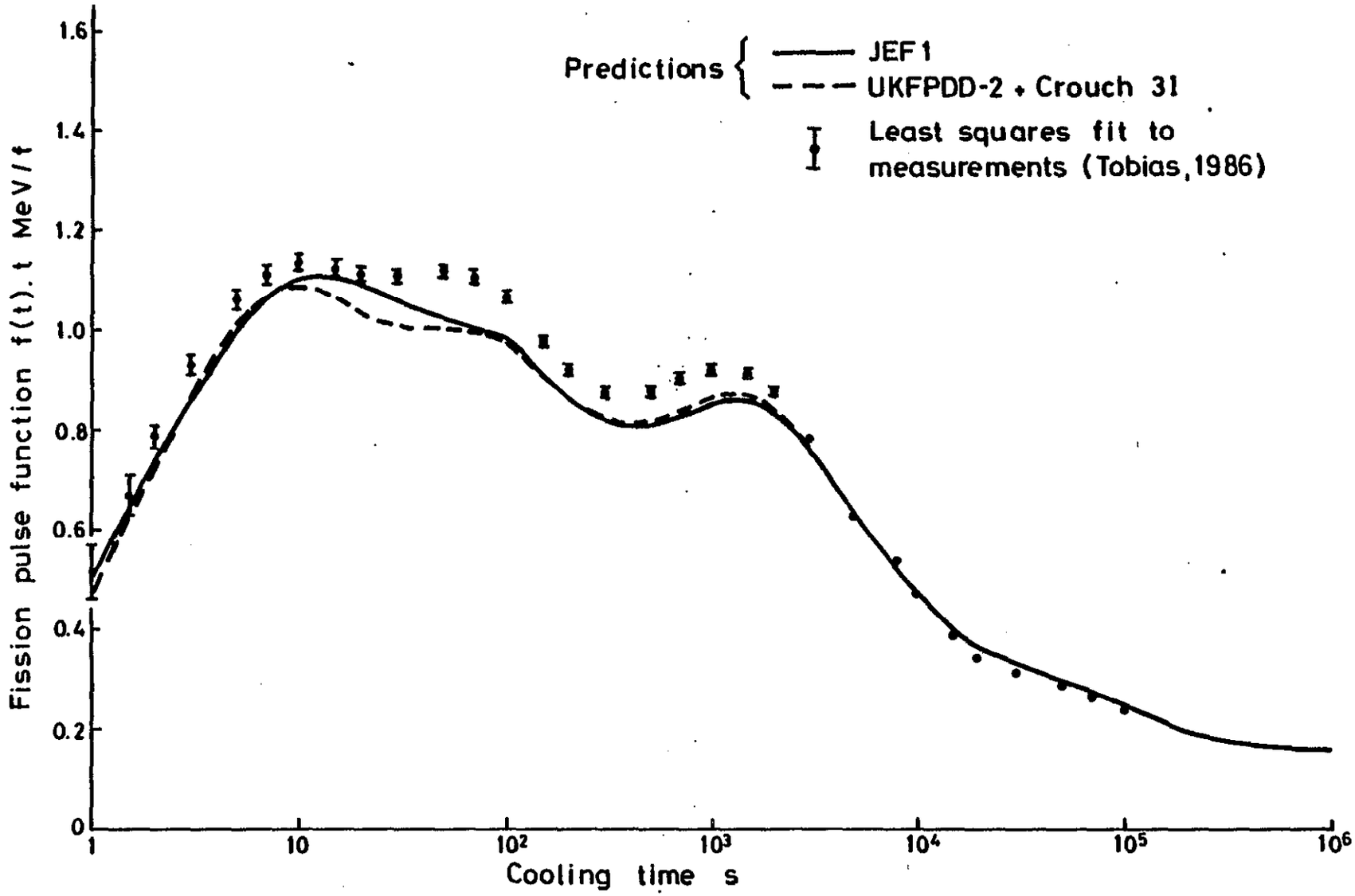


FIG. 10. ^{239}Pu Beta + Gamma Decay Heat - Fission Pulse.

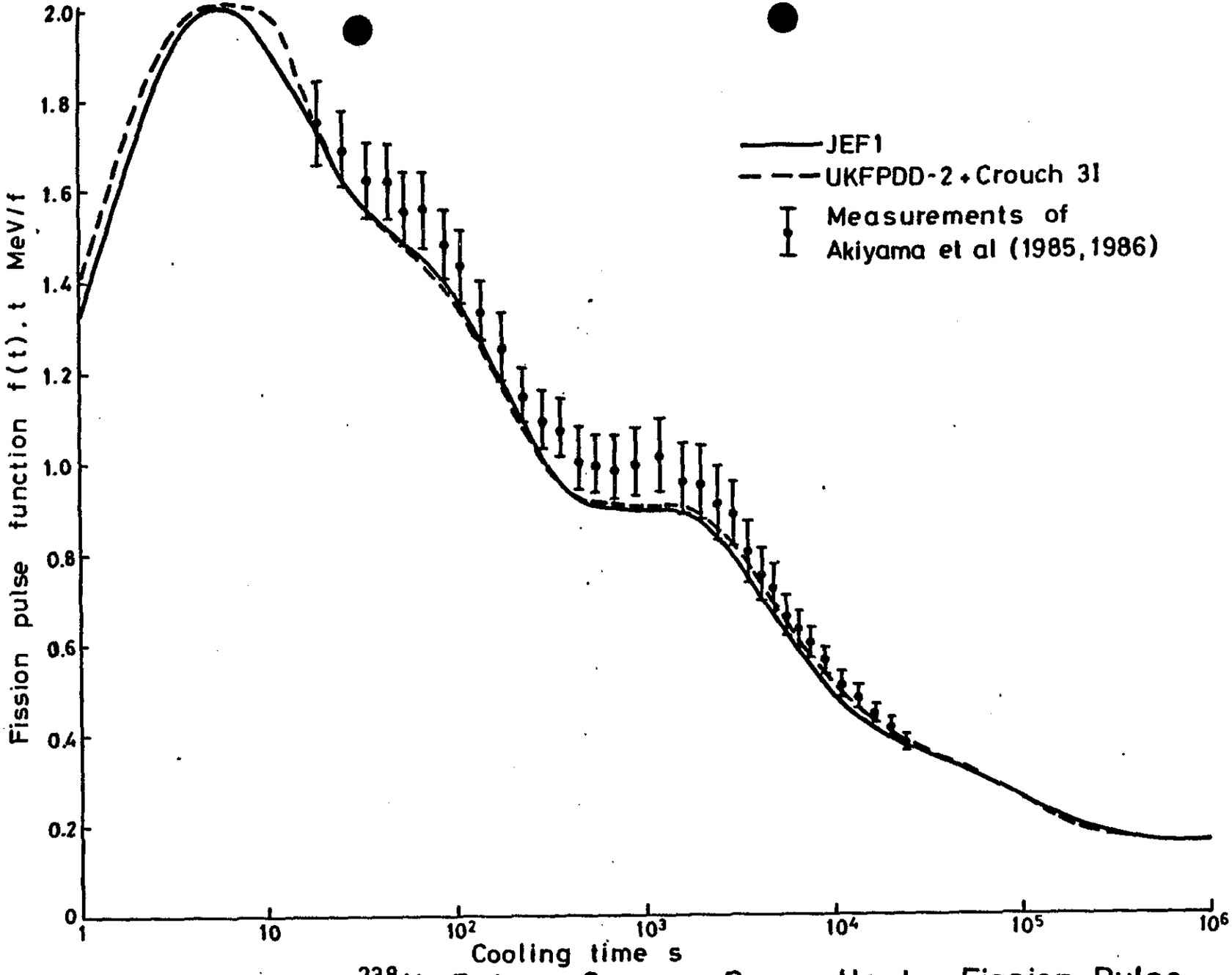


FIG.11. ^{238}U Beta + Gamma Decay Heat - Fission Pulse.

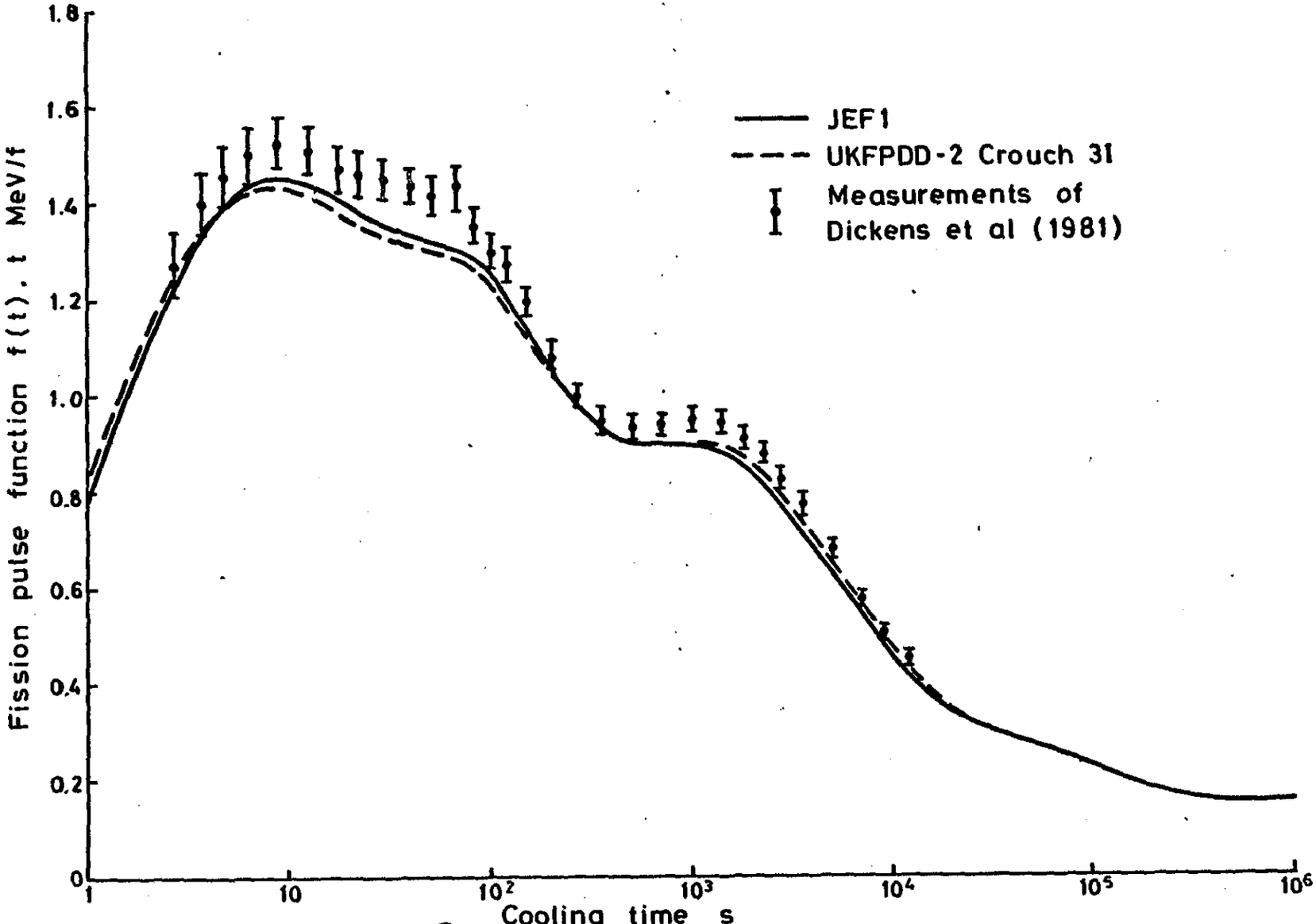


FIG.12. ^{241}Pu Beta + Gamma Decay Heat ● Fission Pulse