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EXPERIMENTAL DETERMINATIONS OF AVERAGE BETA AND GAMMA ENERGIES AND THEIR USE FOR DECAY HEAT PREDICTIONS

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Abstract: Average beta energies have been determined for 112 fission products by direct beta measurements. A similar study of average gamma energies per decay is under way. The measured set of nuclides contributes the major part of the decay heat in nuclear fuel at short cooling times.

(Keyword: Average beta and gamma energies, decay heat)

Introduction

Average beta and gamma energies of individual fission products are important input data for the prediction of the decay heat in nuclear fuel by the summation method. Both average beta and gamma energies can be determined from nuclear decay schemes provided that these are complete. For short-lived nuclides with high disintegration energies the decay is often found to be very complex containing hundreds of gamma-rays. It is then difficult to ascertain whether a decay scheme is complete and that the levels are correctly placed in the scheme. As the beta branches are usually deduced from the decay schemes (established from gamma-ray studies) and not measured directly it follows that erroneous decay schemes will lead to errors in the average beta energy. In order to avoid difficulties of this kind a series of measurements of the energy spectrum of the beta rays emitted from the decaying nuclides has been carried out. The average beta energy is then extracted from the beta spectrum and can be used to replace average beta energies deduced from decay schemes. This approach is independent of the knowledge of the decay scheme. Moreover, the forbiddenness of the various beta branches does not have to be known. The study is an extension of an earlier experiment at the Studsvik Neutron Research Laboratory /1/. Together with that experiment it comprises 121 fission products of half-life between a fraction of a second and a couple of hours covering 50 - 60 % of the beta heat emitted in nuclear fuel at cooling times between 0 and 100 s after stopping a reactor after a long irradiation period. This figure rises to above 90 % if the effect of long-lived nuclides with well known decay schemes is added. This means that the beta part of the decay heat is now well determined experimentally. Only a small fraction of the effect needs be determined from incomplete decay schemes or estimated from extrapolated or theoretically determined beta strength functions.

The corresponding study of the gamma part of the decay heat is under way. The experiments have been carried out, but the analysis of the data is not yet finished.

Experimental determination of average beta energies

The results to be reported here are by-products of an experiment aiming at the determination of the high-energy part of the antineutrino energy spectrum in the vicinity of a nuclear reactor /2/. In this experiment the beta spectra of fission products of high decay energies are determined and then transformed into the corresponding antineutrino spectra. This conversion requires the full energy spectrum from essentially zero energy to the end-point of the hardest beta branch. Having determined the beta spectra it is easy to evaluate the average beta energy, i.e. the quantity of interest for the decay heat problem.

The experiment has been carried out partly at ISOLDE, CERN, and partly at OSIRIS, Studsvik. In both cases the beta spectrum has been measured with a telescope consisting of a thin plastic detector (ΔE-detector) in coincidence with a thick spectrometer of pure germanium. This spectrometer is useful from about 600 keV to at least 15 MeV. It has been carefully studied using monoenergetic electrons from the "Bill" electromagnetic beta spectrometer at Grenoble /3/, and its response function has been studied up to 9 MeV /4/. For the energy range below 600 keV a second spectrometer - a Si(Li)-detector has been used. A further unit is a Ge(Li)-spectrometer for the determination of the composition of the sample using suitable gamma-rays as labels of the components.

The ISOLDE and OSIRIS experiments were run somewhat differently, but the main line is the following. A number of measurements with different timing were carried out for each mass number ascertaining that the number of measurements was equal to or larger than the number of components. This makes it possible to decompose the measured pulse spectra into pulse spectra of the isobaric or non-isobaric (from delayed-neutron emission) components present in the sample. The next step is to convert the pulse spectra into electron spectra using the response functions of the beta spectrometers. Finally, the spectra of the Ge-telescope and the Si(Li)-spectrometer are joined to give the beta spectrum. This is extrapolated both to zero energy and to the end-point of the hardest beta branch in order to yield the complete beta spectrum. Measurements have been carried out for the mass ranges 79 - 100 and 128 - 146. In these mass ranges the set of beta spectra obtained is reasonably complete lacking only components with inconveniently long half-lives (above a few hours).

Resulting average beta energies

Because of space limitations the table of average beta energies determined in the present work cannot be reproduced here. Instead, reference is made to the Proceedings of a specialists' meeting on Data for Decay Heat Predictions held at Studsvik in 1987 /5/. There the results can be compared to average beta energies of the data files ENDF/B-V /6/, JEF1 /7/, and JNDC /8/ as well as the results of the earlier experiment of Studsvik /1/. The trend of the data can be seen from the average of the ratios between beta energies in the files and the corresponding energies determined in the present work. In the averaging procedure the logarithm of the ratio is used in order to give comparable weight to large and small ratios. It turns out that all three files agree closely with the experimental set of data on the average, the mean ratio being 0.997±0.020 for ENDF/B-V, 1.005±0.028 for JEF1, and 0.971±0.024 for JNDC. These averages are obtained from 121 determinations, however, which makes the errors small. The scattering of the points is better described by the standard deviations of the populations. This is found to be 0.27, 0.36, and 0.31 for the three files, respectively. This means, for example, that 70% of the ratios fall between 0.80 and 1.27 for ENDF/B-V. In spite of this fairly large scattering one may conclude that the average beta energies are rather well represented in the files.

Use of the average beta energies in summation calculations

The new set of average beta energies has been entered into a data file which is essentially based on JEF1-data updated with new experimental information from the OSIRIS facility. Those average beta energies which have not been directly measured are the JEF1 ones. Summation calculations have been carried out with this file for the same conditions as regards irradiation and cooling times as used by Johansson in his integral decay heat measurements /9/ in order to allow a detailed comparison with those measurements. This comparison is done by means of Figs. 1 and 2 for the thermal-neutron induced fission of U-235 and Pu-239, respectively. In both cases the agreement is excellent. Hardly any point falls outside of the limits of error.

The uncertainties of the summation calculation are composed of the errors of the decay data entering into the calculation, including the average beta energy, and the errors of the fission yields. They are typically around 2% for U-235 and 3% for Pu-239 (one standard deviation).

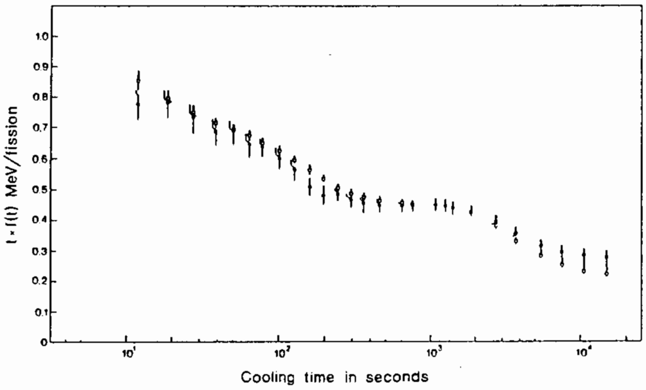

Fig. 1. Beta heat multiplied by cooling time for thermal fission of U-235.
Filled circles: integral measurement /9/
Open circles: summation calculation using new set of average beta energies.
Irradiation time 4 - 120 s.

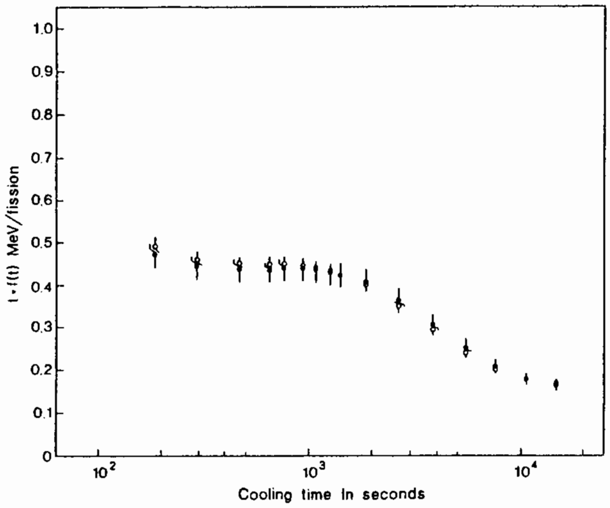

Fig. 2. Beta heat multiplied by cooling time for thermal fission of Pu-239.
Filled circles: integral measurement /9/.
Open circles: summation calculation using new set of average beta energies.

It is also of interest to compare the results using the present library with other summation calculations. This is done in Fig. 3 for U-235. The comparison is done with three curves presented at the Brookhaven meeting in 1983 /10-12/. Apparently, the agreement is next to perfect with the curves based on ENDF/B-V /11/ and the French file /12/, the differences being well within one standard deviation of the calculation. The Japanese file /10/, on the other hand, gives a decay heat curve which is significantly lower than the present calculation for cooling times up to about 500 s.

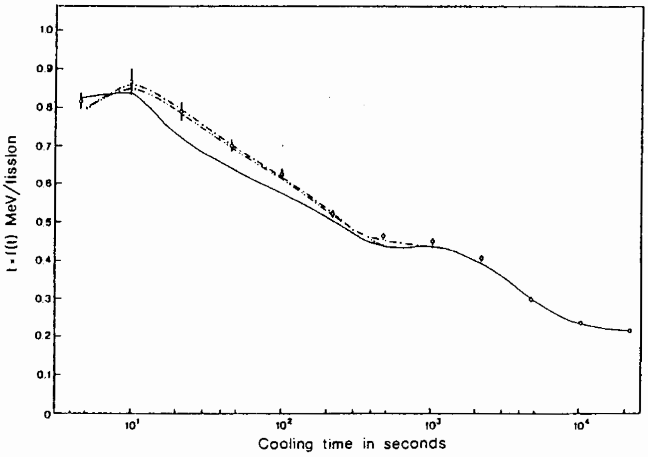

Fig. 3. Beta heat multiplied by cooling time for thermal fission of U-235. Comparison of summation calculations.
Open circles: This work.
Solid line: Ref./10/.
Dash-dot line: Ref. /11/.
Dash-dot-dot line: Ref. /12/.

Average gamma energies

Average gamma energies can be determined in the same manner as the average beta energies. Experimentally, the only difference is that the beta spectrometer system discussed above should be replaced by a suitable gamma spectrometer. Such an experiment has recently been carried at OSIRIS using the same NaI-spectrometer as in ref./9/. This experiment covers the mass ranges 82 - 100 and 130 - 146, thus essentially the same ranges as for the average beta energies.

In the beta case one makes use of the fact that there is one beta particle per decay (with the exception of isomeric states with internal transition branches). This is a very useful information because it means that the average energy of the beta particles is also the average energy emitted per decay. This simplification is not at hand for the average gamma energy. Here the absolute calibration of both the Ge(Li)-spectrometer and the NaI-spectrometer are of utmost importance. Both are needed for the determination of the average gamma energy emitted per decay.

We expect that the combined calibration errors of both spectrometers will lead to an uncertainty of about 3 % in the average energies. This is a systematic error based mainly on the use of calibrated samples (7 samples in all). For each individual nuclide we will then have another error caused by uncertainties in the absolute value of the branching ratios of the gamma-rays used to determine the abundances. This is a systematic error for the nuclide under study, but it may be treated as a statistical error when summing the effect of many nuclides whose gamma branching ratios have been determined independently of each other.

What is the advantage of using the present method to determine the average gamma energy per decay? For nuclides with well-known decay data there is hardly anything to gain. For those nuclides we presumably have a complete list of gamma-rays, with intensities, at hand, and the average gamma energy can be computed in a simple manner. It must be remembered, though, that there is still a systematic error caused by the uncertainty in the determination of branching ratios on the absolute scale. This is precisely the same kind of error which we have in the present treatment. In most cases it will be the main contribution to the uncertainty of the average gamma energy per decay, and both methods should give results of comparable reliability.

For nuclides with incomplete decay data the present method is preferable. It is likely that the deficit is that high-energy gamma-rays have not been measured or resolved with the effect that the average gamma energy obtained by summing the list of gamma-rays will be too low. Our method is not sensitive to such deficiencies.

There exist a number of nuclides for which the decay data are rudimentary comprising only a couple of gamma-rays. For those cases the method given here will yield the first determination of the average gamma energy emitted per decay.

The experiments on average gamma energies are completed, but the analysis is not yet finished, and no results can be presented here.

Another way to determine the average gamma energy $\bar{E}_\gamma$ is to calculate it from the energy balance of the decay using the formula:

$$\bar{E}_\gamma = Q_\beta - \bar{E}_\beta - \bar{E}_\nu - P_n\,(\bar{E}_n + S_n - \sum_i P_{n\gamma i}E_{n\gamma i}) \qquad (1)$$

with

Q_β = total disintegration energy,

$\bar{E}_\beta$ = average beta energy,

$\bar{E}_\nu$ = average neutrino energy,

P_n = delayed-neutron branching ratio,

$\bar{E}_n$ = average energy of the delayed neutrons,

S_n = neutron separation energy (in the daughter nucleus),

$P_{n\gamma i}$ = the probability that the emitted neutron will feed an excited state (i) in the daughter,

$E_{n\gamma i}$ = gamma energy in the decay of this excited state.

Q_β and S_n are taken from the mass table /13/, from Table of Isotopes /14/, from recent and as yet unpublished experimental results at OSIRIS, or determined in the present work. $\bar{E}_\beta$ are from the present work and $\bar{E}_\nu$ from ref. /2/. P_n is taken from a recent compilation by Lund /15/, and $\bar{E}_n$ is obtained from the compilation in Ref./16/. For $P_{n\gamma i}$ and $E_{n\gamma i}$ only little information exists /17/. As the contribution from the gamma decay in the final nucleus after delayed-neutron emission is usually very small, however, it can be neglected in the absence of experimental information. This will hardly affect the result.

One drawback with the determination of $\bar{E}$ using Eq. (1) is that the uncertainty may become rather large as it is composed of the errors of all the quantities appearing in the formula. Determinations using the method outlined above should therefore be more accurate. Nevertheless, Eq.(1) has been used for the determination of $\bar{E}_\gamma$ for all the nuclides for which the average beta energy has been determined. Lack of space makes it impossible to give the results here, but they are tabulated in the above-mentioned Proceedings /5/, and they can there be compared with values from the files ENDF/B-V, JEF1, and JNDC.

Summary

We now have a reasonably complete set of directly determined average beta energies at our disposal. The beta part of the decay heat determined by the use of this set of data is in excellent agreement with recent integral measurements. The corresponding set of average gamma energies will be available later this year. It will then be possible to compare the gamma part of the decay heat obtained using this new set of data, and also the total decay heat, with integral determinations. This will possibly resolve the discrepancies still present in the field.

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