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INTEGRAL DETERMINATION OF THE BETA AND GAMMA HEAT IN THERMAL-NEUTRON-INDUCED FISSION OF ^{235}U AND ^{239}Pu, AND OF THE GAMMA HEAT IN FAST FISSION OF ^{238}U

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Abstract: Decay gamma energy and beta energy release rates for thermal neutron fission of ^{235}U and ^{239}Pu and the gamma energy release rates from ^{238}U for neutrons of 2.3 MeV have been measured by integral measurement. The gamma-rays and beta-rays were measured separately in different spectroscopic measurements. Absolute values have been obtained for the ^{235}U gamma energy over the time range 10 s to 1500 s and beta-energy from 10 s to 10^4 s and for ^{239}Pu gamma- and beta-energy from 10 s to 10^4 s time, and for ^{238}U gamma energy from 50 s to 10^4 s cooling time. The accuracy of the data is better than 7 %. Summation calculations for the beta-energy release rates from ^{235}U and ^{239}Pu /1/ agree within a few % over the entire time range. Summation calculations /1/ gives syste- matically lower gamma energy at short cooling time. For cooling times around 700 s the discrepancy is about 15 %.

(Keyword: Average beta and gamma energies, decay heat, ^{235}U, ^{238}U, ^{239}Pu)

Introduction

The aim of the measurements has been to obtain accurate data for beta- and gamma- energy release for thermal or fast fission of the most important nuclei for reactor decay heat calculations. The nuclides studied are ^{235}U, ^{238}U, and ^{239}Pu. The same technique was used to measure the gamma and the beta radiation from the fission products of ^{235}U, ^{239}Pu, and ^{238}U. The technique involves irradiation of samples with thermal or fast neutrons (^{238}U). The irradiation times used were short compared to the cooling times. After irradiation of a sample it was transported by a pneumatic system to the beta- or gamma-ray detector where the radioactive decay was followed by measuring up to 16 consecutive spectra.

Samples of 110 mg/cm^2 were used for ^{235}U, 3.5 mg/cm^2 for ^{239}Pu, and 1.8 g/cm^2 for the ^{238}U experiment. Neutrons were produced by a 6 MeV Van de Graff accelerator, fast neutrons by the T(p,n) reaction and thermal neutrons by thermalisation of neutrons from the $^{9}Be(p,n)^{9}B$ reaction. The samples were irradiated for 2, 4, 10, 120, and 870 s, and the gamma- or beta-ray emission from the fission products was followed from about 10 s to 10^4 s. Energy spectra were obtained for betas and for gammas separately in different experiments. The gamma rays from fission products were measured with a NaI detector and the beta particles with a Si(Li) detector. The response functions of the detectors were measured with a number of calibration sources. The response functions were used to unfold the measured pulse distributions. The total energy of the gamma rays and the beta particles as a function of cooling time were obtained by integrating the energy distributions.

The fission rate was obtained from calibrated fission chambers and from gamma spectroscopy using a Ge(Li) detector. An accurate measurement of the beta and gamma ray energy release from decaying fission products involves the production of neutrons of known energy and flux, the knowledge of the isotopic composition and the number of nuclei of the samples, and well defined irradiation, waiting, and cooling times. Furthermore, the gamma and beta energy distributions have to be measured with detectors with known efficiency and energy scale. A detailed discussion of all steps to get the results reported here will be published at a later time.

Experimental method

Neutron source and samples

Thermal neutrons for inducing fission in ^{235}U and ^{239}Pu were produced by a 6 MeV Van de Graff accelerator and a thick target of ^{9}Be. The neutrons were thermalized by a moderator of about 1 m^3 of paraffin. The neutron flux available was 10^8 n/(s cm^2). The ratio of fast to thermal neutron fission in ^{235}U was 0.002 according to Monte Carlo calculations and measurements. The neutron flux was monitored with fission chambers during sample irradiation.

Neutrons for the ^{238}U measurement were produced by the T(p,n) reaction and a target of absorbed tritium. The proton energy was optimised for the neutron production cross section, and the target thickness was thin enough to give a well defined neutron energy of 2300 +-100 keV. Cadmium was used to shield the fissile samples during irradiation to lower the effect of thermal neutron fission of ^{235}U atoms present in the sample. The ^{238}U samples had to be located close to the target in order to get a reasonable fission rate. It was therefore not possible to relate the monitored neutron flux to the number of fission in the sample. The accuracy with that method will be to low because of the anisotropy of the neutron source.

All samples used were 10 mm in diameter but of different thicknesses. The samples for the beta measurement had a thickness of 3.5 mg/cm^2. The isotopes ^{235}U and ^{239}Pu were enriched to 99.3 % and electrodeposited onto 8 mg/cm^2 of titanium. The fissionable material was covered with 4 mg/cm^2 of titanium in order to keep the fission products within the sample. For the ^{239}Pu gamma measurement the same samples were used, but thicker samples of metallic ^{235}U of 110 mg/cm^2 enriched to 93.3 % were used for

the ^{235}U measurement. The amount of ^{239}Pu atoms was obtained from the ratio of the gamma line 128.28 keV from the samples and from a calibrated source of ^{239}Pu. The masses of the samples varied between 3.2 mg and 3.6 mg and were determined with an accuracy of 2.5 %.

The low fission cross section for ^{238}U and the neutron flux available, made it necessary to use thick samples (1.8 g/cm^2). Therefore only gamma-rays could be studied for ^{238}U. Samples of various thicknesses (0.214 mg/cm^2, 1.88 mg/cm^2) and enrichment (0.04 % and 0.4 % ^{235}U) as well as natural uranium have been used in order to study the gamma attenuation in the sample and contribution from thermal neutron fission in ^{235}U. The measurements confirmed calculations showing that the effect of thermal neutron fission of ^{235}U atoms in the ^{238}U samples could be neglected and that corrections for attenuation of gamma in the sample can be made with sufficient accuracy. The ^{235}U and ^{238}U samples used for gamma measurements were capsuled by diffusion tight plastic to avoid losses of fission products. All samples used have been supported by cylinders of polythene for transportation by a pneumatic system to the positions for irradiation and measurement.

Beta ray detection system

The beta measurements were carried out with a Si(Li) detector system consisting of a main transmission detector, 25 mm in diameter and 5 mm thick /2/. The detector system was mounted in a vacuum chamber and cooled by liquid nitrogen. Beta particles from a sample have to penetrate 5 mm of air and a window of 3.5 mg/cm^2 of Mo before reaching the detector. The linearity of the beta detector was checked using conversion electrons from ^{207}Bi and end points of the beta spectra from ^{144}Pr, ^{90}Sr, ^{204}Tl, and ^{106}Rh. The strength of these sources were known to 1.5 % and therefore also used to get the absolute efficiency of the detector.

The shape of the response function was obtained from the ^{207}Bi conversion electrons as described in Ref /2/. The effect of beta particles scattered by surrounding material including air must be considered. Therefore all sources for calibration were capsuled in the same way as the ^{235}U and ^{239}Pu samples. The effect of scattering and absorption of beta particles will therefore be included in the efficiency function for the detector. Measured spectra were unfolded by an rectangularly shaped response function with a tail as determined from the ^{207}Bi measurement. The beta energy distribution from decay of ^{106}Ru have been measured with the Si(Li) spectrometer and a double focusing beta spectrometer. The shape of the spectra obtained with the two systems agreed within 5 % over the energy range 400 keV to 3000 keV (400 keV was the lower limit for the focusing spectrometer). The mean beta-ray energy for the two spectra agreed within 2 %.

Gamma ray detection system

A NaI(Tl) crystal (Φ=12.5 cm and l=12.5 cm) was used to detect the γ-rays from the fission products. The crystal was shielded by 10 cm of lead and 10 cm of paraffin. The size of the collimator was 5 cm in diameter and 15 cm in length and had a conic design to diminish the effect of gammas scattered inside the collimator. The detector energy scale and response function were measured with calibration sources. Altogether, 35 pulse distributions for gamma-ray energies ranging from 59.5 (^{241}Am) to 4071.9 keV (^{49}Ca) were obtained. From the measured response functions a response matrix including 60 energies covering the range 40 keV to 6 MeV was generated by interpolation and extrapolation. A bin width of approximately one third of the FWHM of the photo peak was chosen for the matrix. The energy resolution was 20 % for the lowest energy and 7 % for the highest. The measured gamma pulse spectra were converted to energy distributions by the unfolding code FERDOR /3,4,5/ and the measured response matrix. The mean gamma ray energy from known calibration sources were analysed with an accuracy of 1-2 % (100 keV to 3500 keV)

Fission rate the for ^{238}U measurement

The gamma rays from irradiated ^{238}U samples were followed with the Na(I)- and a Ge(Li)- detector simultaneously. The peak energy and decay curve for prominent lines in the Ge(Li)-spectra were used to identify fission products and to calculate the number of induced fissions in the sample. The calculated gamma activity at the end of irradiation was obtained after correction for attenuation of gamma in the sample, the detector efficiency, the branching, growth and decay during irradiation. The number of fissions can be be obtained from the gamma activity and the fission yield. The accuracy of the result will depend on the accuracy of the fission yield, and the yield depends on the neutron energy. Therefore an experiment with the intention to determine the yield of fission products from fast fission of ^{238}U has been carried out. The primary neutron energy was the same as for the decay heat experiment (2.3 MeV). So far the cumulative fission yields for about 40 nuclides have been obtained in the mass regions 87-107 and 129-151. The results have been used for the ^{238}U decay heat measurement.

Well resolved gamma lines from decay of ^{91m}Y, ^{91}Sr, ^{97m}Nb, ^{99m}Tc, ^{132}I, ^{133}I, ^{140}Ba , and ^{147}Nd were used to obtain the number of fissions for samples irradiated for 120 s and for 870 s. The gamma-ray energy obtained from samples irradiated for shorter cooling times were normalised in overlapping time regions.

The absolute number of fissions was obtained with an accuracy of 4 % as determined from systematic errors of the detector calibration, the peak analysing procedure, branching, and the uncertainty of the cumulative yields used.

Fission rate for the ^{239}Pu and ^{235}U measurements

The fission rate for thermal neutron induced fission of ^{235}U and ^{239}Pu was measured by fission chambers of ^{235}U and ^{239}Pu, respectively, located close to the position of the sample during irradiation. From the count rate of the fission chamber, the known efficiency, the number of fissionable nuclei in the chamber and of the samples, the number of fissions during the irradiation was obtained with an accuracy of 3 %. Also the technique of gamma ray spectroscopy as described above was used. For that purpose a sample of ^{239}Pu was irradiated for 15 minutes, and the gamma intensity from the fission products was followed

for more than 100 days. Only fission products with yield known to better than 3 % have been used . The standard deviation of the mean value of the fission rate from the gamma ray spectroscopy method was 2 % and the total uncertainty of the number of fissions was 4 %. The fission chamber was calibrated with an accuracy of 3 % and the result of the two methods agreed within 4 %. The mean value of the two results was used to obtain the number of fissions in the ^{239}Pu samples.

Result of the ^{238}U decay heat measurement

The result of the ^{238}U decay heat experiment is given in Fig 1. Also shown in the figure is the measurement reported in Ref /6/. Individual data points are given without any adjustment to account for finite irradiation time and counting time and plotted versus the average cooling time = waiting time + half of the irradiation time and measuring time. Such an adjustment will lower the product of cooling time and energy by less than 3 % for the shortest cooling time.

Few measurements on ^{238}U have been reported. The Tokyo measurement /6/ was performed at a higher neutron energy (14 MeV). The two curves in Fig. 1 agree within the limits of the errors, but the different neutron energy used for the two experiments make a comparison difficult. Also shown in Fig. 1 is a summation calculation /1/(using the data library FPLIB7A) which give significantly lower γ-energy for a cooling time shorter than about 700 s.

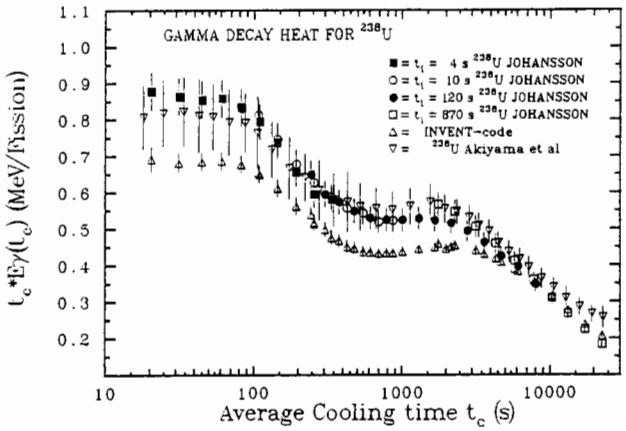

Fig. 1. Gamma heat multiplied by cooling time for fast (2.3 MeV) neutron induced fission of ^{238}U. The data by Akiyama /7/ were obtained at 14 MeV neutron energy and 1000 s irradiation time.

Result of the ^{235}U and ^{239}Pu gamma decay heat measurements.

The gamma energy release after thermal fission of ^{235}U and ^{239}Pu /7/ is shown in Figs. 2 and 3. The experimental data points are presented in the same way as the data from ^{238}U. The results are compared with data from Dickens /8,9/ (thermal neutron energy and short irradiation time), Akiyama /6/ (14 MeV neutron energy and 1000 s irradiation time) and with summation calculations from this laboratory (FPLIB7A) /1/. The experimental data for ^{239}Pu are in excellent agreement but calculated energies are about 15 % lower than the experimental results. Also for ^{235}U the calculated energies are systematically lower except for short (around 20 s) and long cooling time (above 2000 s).

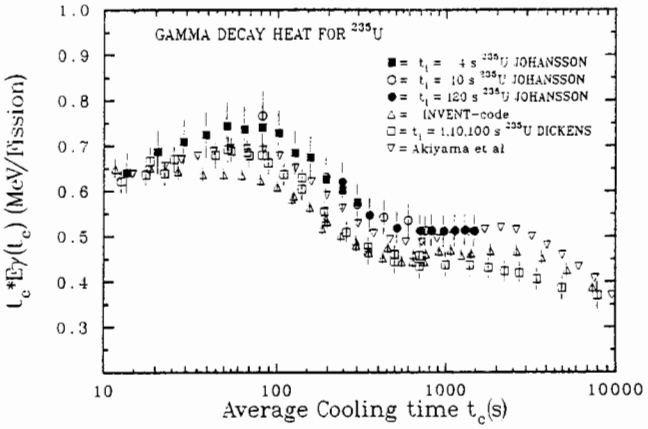

Fig. 2. Gamma heat multiplied by cooling time for thermal fission of ^{235}U. Johansson /7/ , calculation (FPLIB7A library) /1/ and Dickens' experimental data /8,9/.

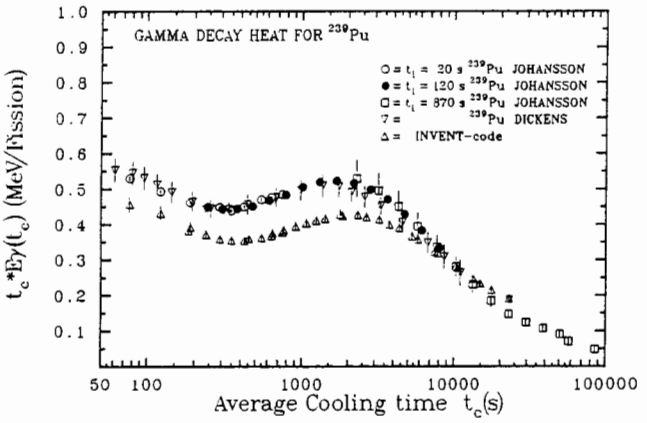

Fig. 3. Gamma heat multiplied by cooling time for thermal fission of ^{239}Pu. Johansson /7/, summation calculation (FPLIB7A library) and Dickens' experimental data /8,9/.

Result of the ^{235}U and ^{239}Pu beta decay heat measurement

The results of the beta ray energy measurements after thermal fission of ^{235}U and ^{239}Pu are given in Figs. 4 and 5 . For comparison experimental data obtained by Dicken's is also shown /8,9/. The origin of the large error bars (Fig. 4) on the present data for long cooling cooling time is small impurities of manganese in the sample capsule, which were not present in the dummy sample used for background measurement. Natural manganese is activated by thermal neutrons giving ^{56}Mn of half life 2.6 h. An estimated contribution of 1 % of the measured beta energy for the cooling time 1000 s will increase to about 20 % at 10000 s. The beta spectra from an irradiated capsule will be measured at a later, time and then the results can be corrected. The calculated values /1/ also shown in Fig. 4 and in Fig. 5 are in excellent agreement with the experimental results.

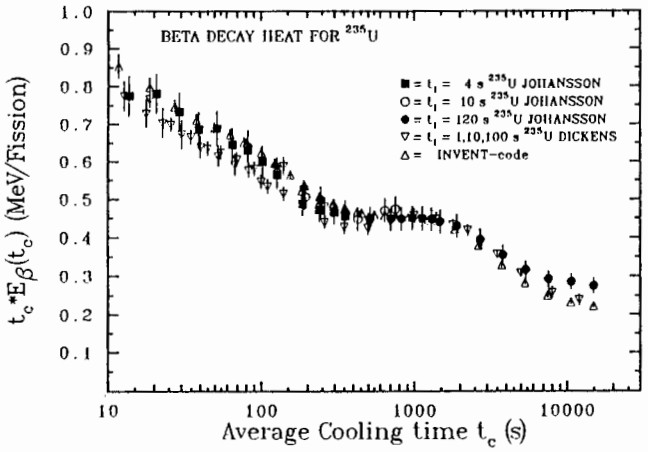

Fig. 4. Beta heat multiplied by the cooling time for thermal fission of ^{235}U. Johansson /1/ , calculation (FPLIB7A library) /1/, Dickens' experimental data /8,9/.

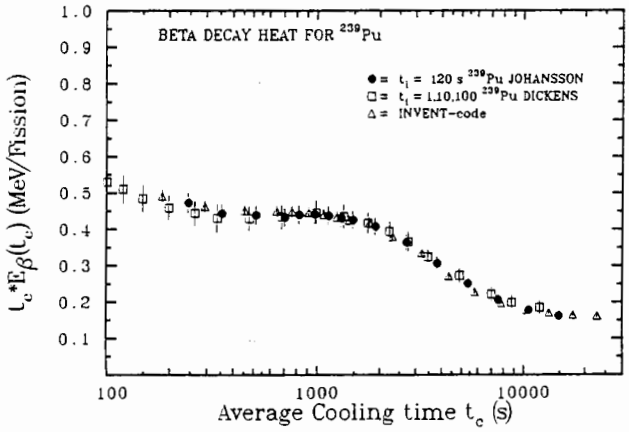

Fig. 5. Beta heat multiplied by the cooling time for thermal fission of ^{239}Pu. Johansson /1/, calculation (FPLIB7A library), Dickens' experimental data /8,9/.

Total decay heat for ^{235}U and ^{239}Pu

The beta and gamma energy release from thermal fission of ^{235}U were added for comparison with the present ANS 5.1 standard /10/ (Fig. 6). The result for ^{239}Pu is shown in Fig. 7. The energies are not adjusted for infinite irradiation time and counting time. Such an adjustment will lower the data points around 300 s cooling time by 1 to 3 % and around 1500 s cooling time for ^{239}Pu (Fig. 7). The circles represent a least squares fit to experimental data available before the Studsvik conference /11/.

Summary and conclusions

Beta and gamma energy release for thermal neutron induced fission of ^{235}U and ^{239}Pu have been measured and compared with Dickens' data /8,9/ and with summation calculations (FPLIB7A) /1/. The measured beta energies and the summation calculation are in excellent agreement for the cooling times studied (10 s-1500 s for ^{235}U and 200 s -10000 s for ^{239}Pu). The results of the gamma measurements agree very well with Dickens' for ^{239}Pu but not for ^{235}U. The calculated /1/ energies are systematically too low (10-20 %) for ^{235}U for ^{239}Pu and for ^{238}U for cooling times around 700 s.

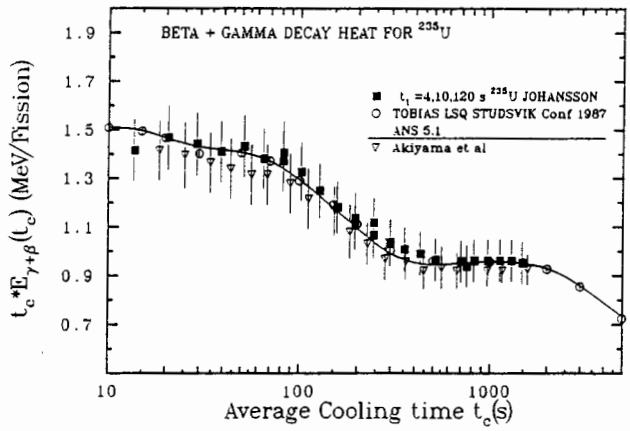

Fig. 6. The total (gamma+beta) energy release rate after thermal fission of ^{235}U. Comparison of the result from this work and /1/ to ANS 5.1 /10/ and Tobias LSQ data from Ref /11/.

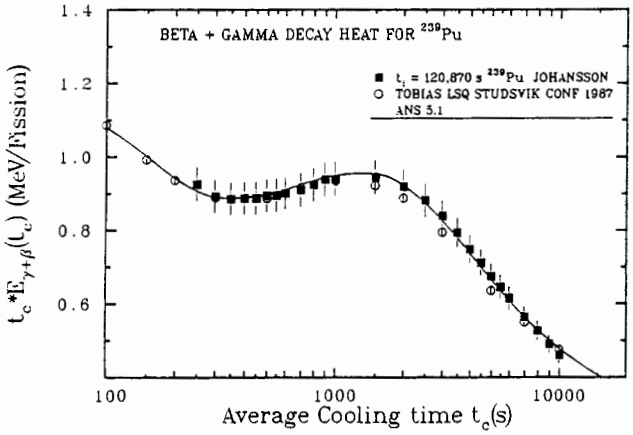

Fig. 7 The total (gamma+beta) energy release rate after thermal fission of ^{239}Pu. Comparison of the result from this work and /1/ to ANS 5.1 /10/ and Tobias LSQ data from Ref /11/.

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