DETERMINATION OF THE ANTINEUTRINO SPECTRUM
FROM ^{235}U THERMAL NEUTRON FISSION PRODUCTS UP TO 9.5 MeV

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A measurement of the cumulated beta spectrum from ^{235}U fission products was performed with a magnetic spectrometer. The energy range 2.0–9.5 MeV was covered achieving a sensitivity of 3×10^{-6} betas per MeV per fission in the high energy region. The absolute rates were calibrated with a precision of 3%. The beta spectrum was converted into the correlated $\bar{\nu}_e$ spectrum to provide a basis for antineutrino experiments at nuclear reactors.

A variety of neutrino oscillation experiments have been carried out recently using nuclear reactors as intense sources of electron antineutrinos. These studies have demonstrated the importance of a knowledge of the $\bar{\nu}_e$ source spectrum even for cases where the search for neutrino oscillations was performed by $\bar{\nu}_e$ detector measurements at several distances from the reactor core [1,2]. The aim of the present work was to further improve the experimental determination of the $\bar{\nu}_e$ source spectrum of reactors both in precision of the absolute rates and at high energies.

Reactor $\bar{\nu}_e$ are emitted with energies up to ~ 10 MeV from the many beta-decaying fission products in the reactor core and approximately 6 $\bar{\nu}_e$ result from each fission event. For power reactors the dominant contributions to the spectrum stem from the thermal neutron induced fission of ^{235}U and ^{239}Pu. Contributions from the fission of ^{238}U and ^{241}Pu are below 10% of the total fission rate.

The source spectrum $N_\nu(0)$ of a nuclear reactor can be derived from a measurement of the cumulated beta spectra of fission products of the fissile isotopes mentioned above. The $\bar{\nu}_e$ spectrum $N_\nu(0)$ is correlated with this beta spectrum by the energy conservation law for individual beta decay branches $E_\nu + E_\beta = E_0$, where E_0 denotes the end point energy of the branch. The feasibility of this method was demonstrated in our earlier work on $\bar{\nu}_e$ spectra from ^{235}U and ^{239}Pu fission [3,4].

These $N_\nu(0)$ spectra have been used in conjunction with the results of recent $\bar{\nu}_e$ detector measurements in the search for evidence of possible neutrino oscillations. Subsequently it has become evident that the absolute precision and the energy range of the available $\bar{\nu}_e$ source spectrum of ^{235}U fission were major constraints for the data interpretation. Thus a new series of measurements on the beta spectrum from ^{235}U fission products has been performed with the magnetic beta spectrometer BILL [5] at the High Flux Reactor of the ILL, with the introduction of several major improvements, namely:

(i) Measurements at reduced (4 MW) and full (57 MW) reactor power for the beta spectrum and efficiency calibration, respectively.

(ii) Detection of the electrons in 115 mm of the declined focal plane of the spectrometer by 32 separate proportional counters of 3.2 mm entrance slit width, defining the momentum resolution as $\Delta p/p = 3.5 \times 10^{-4}$. After transmission through such a wire chamber the electron entered a rearmounted plastic scintillator operated in coincidence to reject background electrons of low energy. Both single and coincidence events were recorded.

(iii) Intensity calibrations now accessible with more

precise neutron capture cross section standards over the whole energy range.

The beta spectrum of ^{235}U fission products was measured at the reduced reactor power of 4 MW in order to improve the signal to background ratio of the measured spectrum. This improvement is mainly due to the increased target mass of ^{235}U permitted at the in-pile target site of the spectrometer (fission heating puts an upper limit on the mass) when measuring at 4 MW. Thus relative to full power, the beta spectrum was kept at a similar intensity whilst the reactor correlated background was significantly reduced. An improvement factor of approximately 10 was in fact achieved. The signal to background ratio was 6, 2.5, 0.5 and 0.12 at 3, 6, 8 and 9 MeV, respectively. The maximum counting rate was 10^3 counts per wire and second.

The target consisted of a 1 mg/cm^2 layer of 93% enriched ^{235}UO$_2$ contained between two Ni foils of 7 mg/cm^2 thickness. The surface area of the layer was 3×6 cm^2. In the thermal neutron flux at the target site ^{235}U undergoes fission. The fission products are stopped in the Ni foils whilst the electrons from the beta decaying fission products are emitted from the target. After a flight path of 13 m the electrons are analysed in the double focussing iron-core spectrometer and recorded in the focal plane using the background rejecting multiwire electron detector. Data points were taken in steps of 50 keV over an energy range of 1.5 to 10.5 MeV with counting times from 30 to 120 s. Three runs were performed covering in total a measuring time of 15 h. This exposure time of ^{235}U to neutrons should bring all beta activities above 3 MeV into equilibrium with the feeding through fission. The underlying background from the beam tube and target material was determined by the same sequence of measurements but with a target of depleted ^{238}U(negligible (n_{th}, f) cross section) designed to have exactly the same geometry and mass as the ^{235}U target within better than 0.3%.

The absolute calibration per fission of the N_β spectrum was based on the proportionality of the reaction rate in the target to $\sigma_n \cdot \phi_n \cdot m$, where σ_n denotes the relevant thermal neutron capture cross section, ϕ_n the neutron flux and m the mass of the isotope (see refs. [3,4]).

For ^{235}U a σ(n, f) cross section of 566(3) b was taken including the non $1/v$ correction for 40°C neutrons of thermal distribution [6]. The isotopic mass of

the ^{235}U target was determined by mass spectroscopy with a precision of 0.5% (CEA, Bruyère-le-Châtel). Masses of other targets (see below) were determined by weighing.

The 4 MW run included a calibration with a metal target of 16.9 mg natural Indium evaporated on an Al backing. Neutron capture produces the beta-active isomers ^{116,116m}In with half lifes of 14 s and 54 min. The K internal conversion electrons (ICE) of the 1.29 MeV, E2 transition in ^{116}Sn was measured in saturation. The partial n_{th} capture cross section for this line is given by $\sigma(n_{th}, \gamma) \times$ ICC. The cross section $\sigma(n_{th}, \gamma)$ for this transition can be evaluated from ref. [7] as 140.4(30) b. A theoretical internal conversion coefficient (ICC) of α_K (E2, $Z = 50$, $E = 1.29$ MeV) = 6.47(7) $\times 10^{-4}$ was adopted [8]. A total accuracy of 3.1% (90% CL) was obtained for this calibration point.

In total the 4 MW run lasted for 46 h. The neutron flux was monitored and was found to vary by less than 1%.

A high energy calibration point was difficult to obtain at 4 MW reactor power, since the spectrometer sensitivity and available measuring time were not sufficient. Thus this calibration was carried out at the full reactor power of 57 MW. Firstly the above mentioned 1.29 MeV line in ^{116}Sn was measured at 4 and 57 MW reactor power. The neutron flux ratio was determined directly by the intensity ratio of this ICE line, with a precision of 1.3%.

The sensitivity gain with full reactor power allowed us to use the ^{207}Pb(n_{th}, γ) ^{208}Pb reaction with $\sigma = 0.712(10)$ b [6] as part of the calibration. The K ICE line of the 7.37 MeV, E1 transition of 100% per neutron capture was measured using a metal target of 34 mg Pb, enriched to 91.6% in ^{207}Pb. A theoretical ICC of α_K (E1, $Z = 82$, $E = 7.37$ MeV) = 9.25(9) $\times 10^{-5}$ b was adopted [8]. A total accuracy of 3.6% was obtained for this calibration point.

In addition the relative response of the spectrometer was determined in a measurement of the ^{113}Cd$(n_{th},$ e$^-)^{114}$Cd reaction, which provides in the range 0.5 to 9 MeV several ICE lines from transitions of known multipolarities [9]. Theoretical ICC's were again taken from ref. [8]. The corresponding (n, γ) intensities were determined precisely in a measurement with pair- and Ge(Li) spectrometers [10], using a mixed CdO and Si$_3$N$_4$ target in a graphite container. The relative ^{113}Cd(n, $\gamma)^{114}$Cd intensities were calibrated with

respect to the well-known intensities of ^{14}N(n, γ)^{15}N [11] and ^{12}C(n, γ)^{13}C [12]. Essentially this calibration resulted in a value of 1.074(49) for the ratio of the BILL spectrometer efficiency in the range 5.5 to 9 MeV relative to that in the range 1.2 to 2.5 MeV. This value is consistent with the ratio of 1.093(48) from the In–Pb calibration. Assuming a monotonically increasing efficiency of the spectrometer with energy the final errors were evaluated from the independent In, Pb and Cd measurements, resulting in total uncertainties of 2.8% and 3.1% for the absolute rates at 1.3 and 7.4 MeV, respectively.
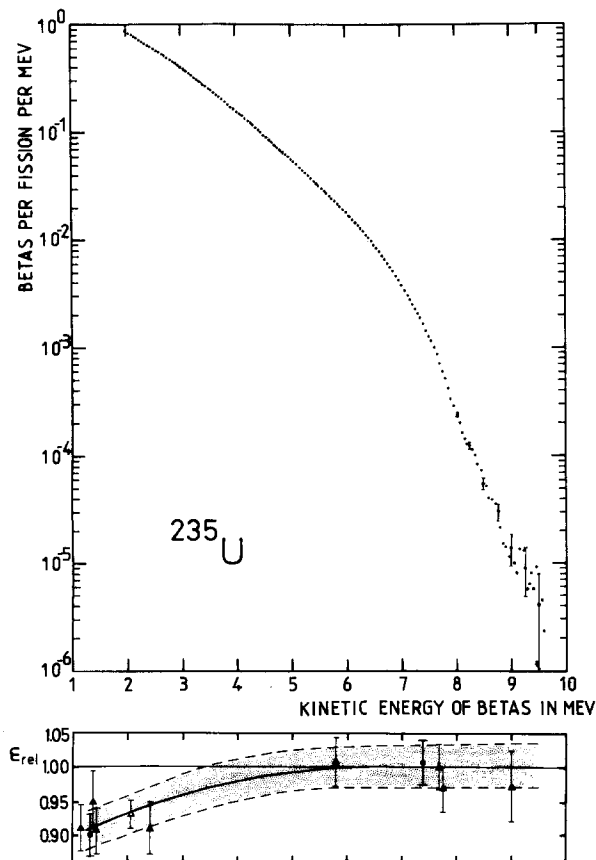
Fig. 1 shows the final N_β spectrum of the ^{235}U fission products. In the lower part of the figure the intensity calibration points are shown. The data are given explicitly in table 1 in bins of 250 keV. At E_β = 3 MeV the difference between the last two ^{235}U runs amounted to 2% indicating that the saturation intensity was nearly achieved. Only the values of the third run (exposure time to neutrons >12 h) were adopted for energies below 5 MeV. It was found that the present result agrees well with our earlier study [+1]. In the high energy region of the N_β spectrum a sensitivity of 3 × 10^{-6} per MeV and fission was obtained, which is an improvement by an order of magnitude. The accuracy in the absolute calibration was improved from our earlier value of 5% to the present 3%. Fig. 2 compares our result with various calculated spectra, demonstrating the difficulty of describing theoretically the beta-decay branches of the many fission products (see also ref. [3]).

The conversion of the experimental beta spectrum into the correlated $\bar{\nu}_e$ spectrum was accomplished by an algorithm similar to that described in ref. [4]. The

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[+1] For comparison the values in ref. [3] have to be increased by 3% since the non 1/v neutron capture cross section of ^{235}U was not taken into account (see also ref. [4]).

Fig. 1. Experimental beta spectrum of ^{235}U fission products. The error bars in the spectrum illustrate the statistical accuracy for bins of 50 keV (90% CL). In the lower part the determined efficiency curve $\epsilon(E_\beta)$ with the uncertainty range (shadowed area) is shown. The errors given for the absolute calibration points (dots) are independent values. For the relative calibration points (triangles) the errors are dominated by correlated uncertainties (see text). The unfilled triangle denotes the 2.1 MeV line in the ^{116m}In decay.
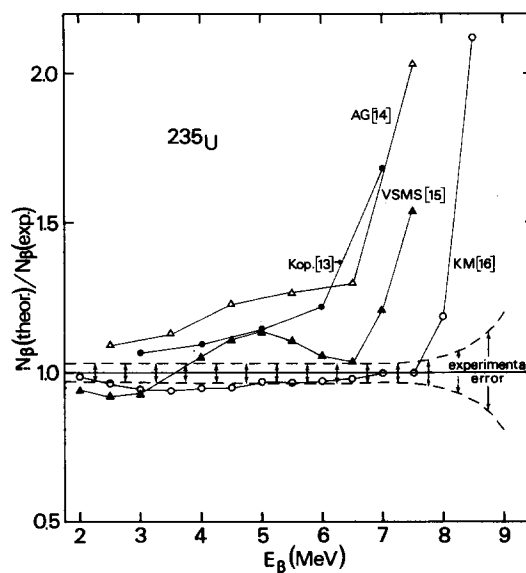

Fig. 2. Ratio between various calculated beta spectra of ^{235}U fission products and the present experimental result. The total uncertainties of the present work are indicated (90% CL).

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Table 1

Experimental beta spectrum from ^{235}U fission products and deduced $\bar{\nu}_e$ spectrum. The exposure time to a constant flux of neutrons was ~12 h.

E (kinetic energy in MeV)	N_β (per fission per MeV)	$\Delta N_\beta(\%)$ statistical error (90% CL)	$N_{\bar{\nu}}$ (per fission per MeV)	$\Delta N_{\bar{\nu}}(\%)$ conversion	total error [a]
2.0	9.36(−1)	<0.5	1.30	3	4.2
2.25	7.71(−1)		1.08		
2.50	6.35(−1)		9.00(−1)		
2.75	5.24(−1)		7.61(−1)		
3.00	4.24(−1)		6.37(−1)		
3.25	3.40(−1)		5.36(−1)		
3.50	2.69(−1)		4.37(−1)		
3.75	2.11(−1)		3.52(−1)		
4.00	1.64(−1)		2.83(−1)		
4.25	1.26(−1)		2.23(−1)		
4.50	9.71(−2)		1.72(−1)		
4.75	7.56(−2)		1.32(−1)		
5.00	5.77(−2)		1.05(−1)		
5.25	4.37(−2)		8.21(−2)		
5.50	3.33(−2)		6.17(−2)		
5.75	2.48(−2)		4.82(−2)		
6.00	1.81(−2)		3.70(−2)	3	4.3
6.25	1.32(−2)		2.70(−2)		
6.50	9.28(−3)	<0.5	2.03(−2)		4.4
6.75	6.17(−3)	0.5	1.50(−2)		
7.00	3.88(−3)	0.6	1.05(−2)	3.6	4.7
7.25	2.36(−3)	0.7	6.68(−3)		
7.50	1.33(−3)	0.9	4.29(−3)	4	5.0
7.75	6.29(−4)	1.5	2.69(−3)		5.4
8.0	2.48(−4)	2.9	1.36(−3)	5	6.0
8.25	1.23(−4)	4.7	4.13(−4) [b]		7.0
8.50	6.16(−5)	7.9	2.37(−4)	6	10
8.75	2.82(−5)	13	1.29(−4)		15
9.00	1.17(−5)	26	5.6 (−5)	7	27
9.25	1.03(−5)	30	2.2 (−5)		31
9.50	4.5 (−6)	55	1.4 (−5)	10	56

[a] Quadratic sum over the uncertainties of the statistical precision, the conversion procedure and the absolute rates (90% CL).
[b] Significant change in shape at this energy: values for $N_\nu \times 10^{-4}$ are 13.6, 8.9, 5.02, 3.24, 2.79 and 2.37 at E_ν = 8.0, 8.1, 8.2, 8.3, 8.4 and 8.5 MeV, respectively.

beta spectrum was approximated by 30 hypothetical beta branches of allowed shapes P_i to obtain the endpoint distribution $\{a_i, E_0^{(i)}\}$:

$$N_\beta = \sum_i a_i \cdot g_i'(E, E_0^{(i)}) \cdot P_i[E, E_0^{(i)}, \bar{Z}(E_0^{(i)})], \qquad (1)$$

with the normalisation

$$\int P_i \, dE = \int g' P_i \, dE . \qquad (2)$$

The radiative correction term g' was included following the theory of Sirlin [17]. The function $\bar{Z}(E_0^{(i)})$ describes the empirical mean proton number of the fission products with beta branches of endpoint energy $E_0^{(i)}$ (from refs. [15,16]) and can be approximated for ^{235}U fission products by:

$$Z_0^{(i)} \approx 49.5 - 0.7 E_\beta - 0.09 E_\beta^2, \quad Z_0 \geqslant 34, \qquad (3)$$

with E_β in MeV. Due to the high statistical accuracy of the present data and the steep slope of the beta

spectrum the endpoint distribution $\{a_i, E_0^{(i)}\}$ according to eq. (1) is well determined and possible variations alter N_ν by not more than 1% apart from statistical fluctuations.

To obtain the N_ν spectrum, each individual, hypothetical branch was converted. The radiative correction was removed since it is small for antineutrinos compared to the betas. The $\bar\nu_e$ spectrum was then obtained from the sum:

$$N_\nu = \sum_i a_i P_i [(E_0^{(i)} - E), E_0^{(i)}, \bar{Z}(E_0^{(i)})] . \qquad (4)$$

A test of our conversion procedure takes advantage of the explicit summing of individual beta branches in calculated spectra. For several calculations Borovoi et al. [18] derived a correlation function between N_ν^{cal} and N_β^{cal} (E_β), which was found to be rather independent of the theoretical approach, but was a strong function of E_β. For relativistic cases, $E_\beta \gg m_e c^2$, a closer correlation between N_ν and N_β can be obtained when the spectra are compared at energies E_ν and $E_\beta + m_e c^2$, respectively.

$$N_\nu(E_\nu) = k(E_{tot}) N_\beta(E_{tot}) ,$$

$$E_{tot} = E_\beta + m_e c^2 . \qquad (5)$$

The function $k(E_{tot})$ is expected to be close to unity. In fig. 3, $k(E)$ is plotted for several theoretical approaches and also for our conversion procedure outlined above. Since no radiative corrections were included in the theoretical spectra we have removed this term for comparison in fig. 3.
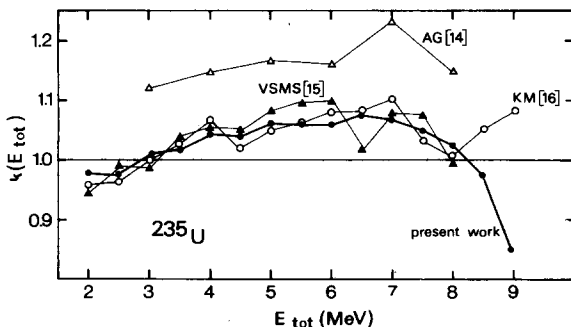

Fig. 3. The function $k(E) = N_\beta(E_{tot})/N_\nu$ for calculated spectra and for our result. For comparison the radiative correction and ΔN^{WC} terms are removed in our conversion procedure, since these terms are omitted in the theoretical spectra.

For cases where the corresponding N_β spectra differ by not more than ~20% the $k(E)$ functions agree within a few percent. For larger differences of the N_β spectra the deviations in $k(E)$ are significant demonstrating the limit of the approximation (5). The present conversion procedure depends on the shape of beta branches used in the decomposition of the experimental spectrum according to eq. (1). In calculated spectra allowed shapes with the usual Z dependent Fermi function are used. We have added in our procedure the first order Z independent radiative corrections [17]. The remaining shape correction terms are due to weak magnetism, higher Coulomb terms and the occurrence of first forbidden transitions in the beta spectrum. According to ref. [18] the influence of first forbidden transition on the conversion remains smaller than 1%. Weak magnetism and higher Coulomb terms become increasingly important for higher E_0 values. An estimate by Vogel [19] leads to a correction function

$$\Delta N_\nu^{WC}(E_\nu) \approx 0.65 \ (E_\nu \ (MeV)-4)\% . \qquad (6)$$

Since this term depends on the matrix element of the individual beta decay a large uncertainty of 100% of the correction was assumed. In table 1 the final N_ν spectrum of ^{235}U fission products is given as derived from our experimental beta spectrum. The conversion error (column 5) is composed of a global error estimated from the discussion of fig. 2, the variation due to the uncertainty of ±1 charge units in the function $Z(E_0^{(i)})$ and the uncertainty in the ΔN_ν^{WC} term. The total error is taken as the quadratic sum of individual errors.

A more detailed presentation of the experiment is in preparation. We are planning to measure the beta spectrum of ^{241}Pu fission products by a similar method to determine this small but strongly varying component in the $\bar\nu_e$ spectrum of a power reactor.

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