ANTINEUTRINO SPECTRA FROM ^{241}Pu AND ^{239}Pu THERMAL NEUTRON FISSION PRODUCTS

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The antineutrino spectrum of fission products from thermal neutron induced fission of ^{241}Pu was derived from a measurement of the correlated beta spectrum. The energy range 1.5 MeV to 9 MeV was covered and a precision of 4% was achieved at 4 MeV. A revised version of the antineutrino spectrum from ^{239}Pu fission is also presented.

A variety of fundamental problems in charged and neutral current reactions with low energy antineutrinos can be investigated with reactor $\bar{\nu}_e$ under the favourable condition of a high source strength [1,2]. A nuclear reactor emits approximately 2×10^{17} $\bar{\nu}_e$ per megawatt of thermal power with energies up to about 10 MeV, stemming from the beta-decay of fission products in the core. For such investigations an accurate knowledge of the emitted $\bar{\nu}_e$ spectrum, both in intensity and in shape, is of major importance as demonstrated by recent neutrino oscillation and inverse beta-decay cross section studies [3].

For a power reactor the dominant contributions to the total antineutrino spectrum stem from the thermal neutron induced fission of ^{235}U and ^{239}Pu. The contributions from ^{238}U and ^{241}Pu are both less than 10%, with the ^{241}Pu component varying strongly as a consequence of breeding. The spectral shapes of these various components are very different and thus the total spectrum alters with the time evolution of the reactor core.

In earlier studies we have determined experimentally the $\bar{\nu}_e$ spectrum $N_{\bar{\nu}}$ from thermal neutron induced fission of ^{235}U and ^{239}Pu [4–6]. In the present paper we present our final results on the $\bar{\nu}_e$ spectrum from ^{241}Pu fission. Some preliminary data, without an absolute calibration were already given in ref. [6]. A revised and improved version of $N_{\bar{\nu}}$ for ^{239}Pu, based on the experimental results presented in ref. [4], is also given here.

The procedure followed to determine the antineutrino spectrum of ^{241}Pu fission products was carried out in two main stages as in our earlier experiments on ^{239}Pu and ^{235}U [4,5]. The cumulated beta spectrum of fission products from a sample of ^{241}Pu exposed to a constant flux of thermal neutrons was measured first. The experimental spectrum was then converted into the correlated antineutrino spectrum $N_{\bar{\nu}}$.

The experiment was performed at the beta spectrometer BILL of the ILL High Flux Reactor [7]. A layer 0.13 mg cm^{-2} of PuO_2, 83% enriched in ^{241}Pu, was evaporated on to a 2×6 cm^2 area of a 7 mg cm^{-2} nickel foil. The PuO_2 layer was then covered by another Ni foil of 7 mg cm^{-2}. The target was exposed to the thermal neutron flux of 3×10^{14} n cm^{-2} s^{-1} at the in-pile target site of the spectrometer. Fission products from the ^{241}Pu (n_{th}, f) reaction were contained by the Ni foils while the beta particles could emerge and were momentum analysed in the 13 m distant spectrometer magnets. A 32 wire proportional counter in coincidence with a rear-mounted

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plastic scintillator recorded the electrons in the focal plane. The energy range of 1–10 MeV was covered, in steps of 50 keV, several times during the 45 hours of exposure to neutrons. The counting rate in the spectrum was ~500 counts per s per wire at 4 MeV. After one day the beta activity was in saturation at least for energies above 2 MeV. For the spectrum below 3.5 MeV only data from the last run (exposure time > 37 h) were adopted.

The background in the measurements came mainly from the Ni foils in the intense radiation field at the target site. It was determined using a target of identical dimensions to those of the ^{241}Pu target, but with the fissile material replaced by a layer of bismuth. The signal to background ratio was measured as 0.8 and 0.1 at 2.5 MeV and 8 MeV, respectively.

To obtain absolute calibrations of the spectra per fission comparative measurements of internal conversion electron (ICE) lines with well-known partial neutron capture cross sections σ_i were performed. Since the reaction rate in the target is proportional to $\sigma_i\phi_n m_i$ (ϕ_n neutron flux, m_i number of target atoms) this method allows a normalization of the beta emission rate per fission without measuring the absolute efficiency of the spectrometer. As in our earlier work [5], ICE calibration lines in the ^{116m}In beta decay, ^{207}Pb(n,e$^-$) and ^{113}Cd(n,e$^-$) were used in order to cover the energy range from 1.2 to 9 MeV. The neutron flux was monitored to a precision of better than 0.5% throughout the measurements. The measured efficiency curve showed an increase of (10±3)% from 1.5 to 7 MeV. The absolute calibration was precise to 2.9% and 3.2% (90% CL) at 1.5 MeV and 7 MeV, respectively. These uncertainties already include those arising from the ^{241}Pu mass and ^{241}Pu(n, f) cross section. The ^{241}Pu mass in the target was determined after the irradiation by mass spectroscopy with an accuracy of 1%. It was found that the target was contaminated with the fissile isotope ^{239}Pu at a level of 6.7%. A value of 1075(9) b was adopted for the ^{241}Pu(n, f) cross section including the non $1/v$ correction for a maxwellian neutron spectrum in the 50°C heavy water moderator of the reactor.

In fig. 1 the measured beta spectrum from ^{241}Pu fission products is shown. The ^{239}Pu contamination has been subtracted using our data from ref. [4]. The data are explicitly given in table 1 in energy bins of 250 keV. A comparison with various theoretical predictions is shown in fig. 2.

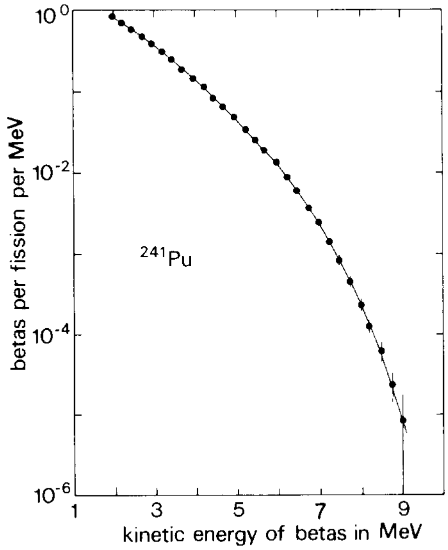

Fig. 1. Experimental beta spectrum of ^{241}Pu fission products. The error bars in the spectrum correspond to the statistical accuracy and variation between the series of measurements for 250 keV bins at 90% CL.

The conversion method from N_β into $N_{\bar\nu}$ has been discussed in detail in refs. [5,6]. Briefly, the experimental beta spectrum is approximated by a set $\{a_i, E_0^{(i)}, \bar{Z}_i(E_0^{(i)})\}$ of hypothetical beta-branches with amplitude a_i, end-point energy $E_0^{(i)}$ and proton number $\bar{Z}_i$. In total thirty allowed beta shapes with radiative correction were taken for this purpose and described the experimental spectrum by better than 1%. The mean proton number $\bar{Z}$ varies with the end-point energy $E_0^{(i)}$ and is taken from semi-empirical data [10]. The antineutrino spectrum is then obtained as the sum of the individually converted branches i. As in ref. [5] the $\bar{\nu}_e$ spectrum was corrected by a few percent for higher Coulomb terms and weak magnetism, following the estimate in ref. [11]. The final $\bar{\nu}_e$ spectrum and its uncertainties are given in table 1. The systematic error in the conversion procedure includes the uncertainties of two units in $\bar{Z}_i$, the variation by using different sets of $\{a_i, E_0^{(i)}\}$, and those associated with the weak magnetism and Coulomb correction terms. In the case of these correction terms an uncertainty of the order of the correction term itself was assumed.

Table 1
Measured beta spectrum N_β from ^{241}Pu fission products for an exposure time to neutrons of 1.8 days and deduced $\bar{\nu}_e$ spectrum $N_{\bar{\nu}}$. Also listed is the $\bar{\nu}_e$ spectrum of ^{239}Pu deduced in a refined way from the beta spectrum in ref. [4]. All errors are given at 90% CL.

E (kinetic energy in MeV)	^{241}Pu					^{239}Pu	
	N_β (per fission per MeV)	ΔN_β (%) [a] statistical error	$N_{\bar{\nu}}$ (per fission per MeV)	$\Delta N_{\bar{\nu}}$ (%) conversion	total error [b]	$N_{\bar{\nu}}$ [a] (per fission per MeV)	$\Delta N_{\bar{\nu}}$ (%) total error [c]
1.50	1.31	<1.0	1.56	4	5.0	1.45	5.2
1.75	1.10		1.42			1.26	
2.00	9.1(−1)	<1.0	1.24		4.3	1.07	4.5
2.25	7.5(−1)		1.06			8.9(−1)	
2.50	6.19(−1)	<1.0	8.7(−1)	2.5	4.0	7.1(−1)	4.3
2.75	5.05(−1)		7.5(−1)			5.99(−1)	
3.00	4.07(−1)	1.0	6.23(−1)		4.0	4.91(−1)	4.3
3.25	3.22(−1)		5.20(−1)			3.97(−1)	
3.50	2.54(−1)	1.2	4.20(−1)		3.9	3.17(−1)	4.3
3.75	1.97(−1)		3.34(−1)			2.48(−1)	
4.00	1.50(−1)	1.5	2.70(−1)	2	3.9	1.90(−1)	4.4
4.25	1.12(−1)		2.10(−1)			1.48(−1)	
4.50	8.5(−1)	1.9	1.57(−1)		4.2	1.07(−1)	4.8
4.75	6.40(−2)		1.18(−1)			7.9(−2)	
5.00	4.75(−2)	2.1	9.2(−2)		4.4	5.76(−2)	5.2
5.25	3.48(−2)		6.96(−2)			4.41(−2)	
5.50	2.50(−2)	3.0	5.25(−2)	2.5	4.9	3.50(−2)	5.9
5.75	1.77(−2)		3.82(−2)			2.66(−2)	
6.00	1.24(−2)	3.9	2.76(−2)		5.6	1.77(−2)	6.8
6.25	8.8(−3)		1.89(−2)			1.26(−2)	
6.50	6.00(−3)	4.5	1.39(−2)		6.1	9.4(−3)	7.4
6.75	3.89(−3)		1.01(−2)			6.94(−3)	
7.00	2.37(−3)	5.5	6.83(−3)	3	7	4.68(−3)	11
7.25	1.43(−3)		4.11(−3)			3.05(−3)	16
7.50	8.4(−4)	6	2.54(−3)		8	1.80(−3)	19
7.75	4.58(−4)		1.59(−3)			8.8(−4)	27
8.00	2.28(−4)	9	8.9(−4)	5	11	5.0(−4)	35
8.25	1.18(−4)		4.36(−4)		17	3.5(−4)	50
8.50	5.5(−5)	20	2.35(−4)		24	2.2(−4)	80
8.75	2.4(−5)	37	1.20(−4)		38		
9.00	9.(−6)	90	4.70(−5)		90		

[a] Includes the mean square variation within the series of the measurements.
[b] Quadratic sum over the statistical, conversion and absolute calibration errors. The latter is 2.9% and 3.2% at 1.5 MeV and 7 MeV, respectively.
[c] Quadratic sum over all errors. The conversion error is similar to that of ^{241}Pu. For the other uncertainties see ref. [4].

The conversion procedure was finally tested by comparison of the ratio $N_{\bar{\nu}}(E_{\bar{\nu}})/N_\beta(E_{tot})$, $E_{tot}=E_\beta+m_e c^2$, with theoretical spectra, where individual branches are summed together. This ratio must be close to unity and similar for all approaches (see ref. [5]). The ratios of our data to those in refs. [2,9] were found to agree within a few percent.

Since the radiative and higher order correction terms were not included in our earlier work on ^{239}Pu, the revised $N_{\bar{\nu}}$ for ^{239}Pu deduced from the experimental beta spectrum in ref. [4] has been added to table 1. Compared to ref. [4] the values for $N_{\bar{\nu}}$ are a few percent higher at elevated energies, but within the error range quoted therein.

With the present data and those in ref. [5], the major part of the antineutrino spectrum of a reactor can now be constructed, where in general the fission rates for the individual isotopes are well-known parame-

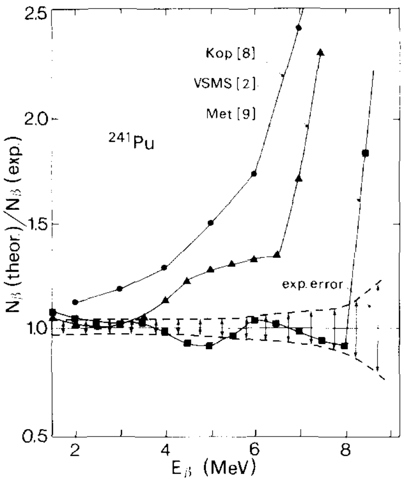

Fig. 2. Ratios of various calculated beta spectra of ^{241}Pu fission products to the present spectrum. The total absolute uncertainty of the present work is indicated (90% CL).

ters for a specific reactor core. However, for $\bar{\nu}_e$ from the fast neutron induced fission of ^{238}U in power reactors one has still to rely on theoretical predictions. A possible experiment on ^{238}U would be a comparative measurement of the beta spectra from ^{238}U(n_{fast}, f) and ^{235}U(n_{th}, f) using the same beta detector. In comparison with the precise ^{235}U data [5] the response function of the detector system can be determined. The absolute rates could be calibrated off-line via long living γ-activities of fission products for which the fission yield is well known in both reactions. The ultimate limit to our knowledge of the $\bar{\nu}_e$ spectra remains the N_β to N_ν conversion procedure, although the uncertainties given here include uncertainties in the spectral shape and the errors are in general smaller in evaluating integral antineutrino cross sections.

As an example the integral cross section for the reaction $\sigma(\bar{\nu}_e+p\to n+e^+)$, being proportional to $(E_{\bar{\nu}}-1.8\text{ MeV})^2/\tau_n$, can be predicted with the present data for the different contributions to the reactor neutrino spectrum with a precision of 3.2%, 3.9% and 3.6% (90% CL) for ^{235}U, ^{239}Pu and ^{241}Pu fission, respectively. The error for the neutron lifetime $\tau_n=896(10)$ [12] is not yet included. The conversion error from the measured beta spectra to the antineutrino spectra enters here only at a level of 1%, governed by the uncertainty in Z of two units.

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