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A study of antineutrino spectra from spent nuclear fuel at Daya Bay*

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Abstract: The Daya Bay Reactor Antineutrino Experiment is designed to determine the as yet unknown neutrino mixing angle, θ_{13}, by measuring the disappearance of electron antineutrinos from several nuclear reactor cores. The projected sensitivity in $\sin^2(2\theta_{13})$ of better than 0.01 at a 90% CL should be achieved after three years of data-taking. Antineutrinos emitted from spent nuclear fuel (SNF) distort the soft part of the energy spectrum. In this article, a calculation of the antineutrino spectra from the long-life isotopes in SNF is performed. A non-equilibrium generation of long half-life isotopes during the running time of the reactor is also analyzed. Finally, we show that the antineutrino event rate contribution from SNF, which has been stored in the SNF pool for several years, may be non-negligible.

Key words: spent nuclear fuel, antineutrino spectrum, non-equilibrium, event rate

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1 Introduction

The Daya Bay Antineutrino Oscillation Experiment [1] was designed to detect the $\overline{\nu}_e$ emitted from the reactor via the inverse beta decay reaction:

$$\overline{\nu}_e + p \longrightarrow e^+ + n. \quad (1)$$

The measured quantity is the survival probability for $\overline{\nu}_e \longrightarrow \overline{\nu}_e$, which is given by

$$P \approx 1 - \sin^2(2\theta_{13}) \sin^2\left(\frac{1.27\Delta m^2_{13} L}{E}\right), \quad (2)$$

where the baseline $L \approx 2$ km. The goal of the experiment is to determine $\sin^2(2\theta_{13})$ at a sensitivity of 0.01 or better at a 90% CL after three years of data-taking.

To achieve the initial goal, the control of systematic uncertainties is critical. Refs. [2–6] show that the antineutrinos from spent nuclear fuel (SNF) can introduce unwanted distortion in measured spectra and thus influence the interpretation of data analysis in reactor-based neutrino oscillation experiments. In this article, a calculation of antineutrino spectra from SNF for a typical reactor, such as Daya Bay, is performed and a non-equilibrium generation of long half-life isotopes during the running of the reactor is also obtained. Combining with the cross section of the reverse beta decay reaction, the contribution of the non-equilibrium antineutrino event rate is estimated. It should be noted that in this calculation, we assume that the reactor is running for 330 days for a full cycle, without stopping to change fuel in between, and then all the fuels are removed to the SNF storage pool after the cycle.

2 The chosen isotopes

As the threshold of the reverse beta decay reaction is around 1.8 MeV, and the isotopes with a short half-life will reach equilibrium right after the reactor on or decay rapidly after being taken out of the reactor, the chosen isotopes should hold $E_d \geqslant 1.8$ MeV, $T_{1/2} \geqslant 10$ h. All these isotopes are listed in Table 1.

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Table 1. Isotopes from fission fragments with $E_{\rm d} \geqslant$1.8 MeV, $T_{1/2} \geqslant$10 h.

M	$T_{1/2}$	E_0/MeV	D	$T_{1/2}$	E_0/MeV
^{90}Sr	28.78 a	0.546	Y	64.1 h	2.282
^{91}Sr	9.63 h	2.699	Y	58.51 d	1.544
^{93}Y	10.18 h	2.874	Zr	1.53×10^6 a	0.091
^{97}Zr	16.9 h	2.658	Nb	72.1 min	1.934
^{106}Ru	373.6 d	0.039	Rh	29.8 s	3.541
^{112}Pd	21.03 h	0.288	Ag	3.13 h	3.956
^{125}Sn	9.64 d	2.364	Sb	2.758 a	0.767
^{131m}Te	30 h	0.182	Te	25 min	2.233
^{132}Te	3.204 d	0.493	I	2.295 h	3.577
^{159}Sm	9.4 h	0.722	Eu	15.19 d	2.451
^{140}Ba	12.75 d	1.047	La	1.678 d	3.762
^{144}Ce	284.9 d	0.319	Pr	17.28 min	2.997

3 Calculation of the activity

In a running reactor, the fission fragment can be generated in several ways, including fission of the nuclear fuel and decay from the mother nucleus and (n,γ) reaction. At the same time, because of their own radioactivity and exposure in a high flux neutron field, the fragments vanish via self-decay and the (n,γ) reaction. The calculation can be divided into three steps.

3.1 Generation from fission

The following differential equation was used:

$$\frac{{\rm d}N(t)}{{\rm d}t} = \sum_i^{\rm isotopes} f_i(t)Y_i - \lambda N(t) - \sum_j \sigma(E_j)\phi(E_j)N(t), \tag{3}$$

where Y_i is the cumulative yield from a certain nuclear fuel (^{235}U, ^{239}Pu, ^{240}Pu, ^{241}Pu and ^{242}Pu) and σ is the cross section of the (n,γ) reaction; the data used were obtained from ENDF/B-VII [7]. The cumulative yield of ^{238}U was obtained from Ref. [8], $f(t)$ is the fission rate of the fuel as a function of reactor running time and $\phi(E)$ is the neutron flux in the core, including two groups of data (0.625 eV, 100 keV).

3.2 Generation from decay

The following equation group was taken:

$$\begin{cases} \dfrac{{\rm d}N_1(t)}{{\rm d}t} = \sum\limits_i^{\rm isotopes} f_i(t)Y_i - \lambda_1 N_1(t) - \sum\limits_j \sigma_1(E_j)\phi(E_j)N_1(t) \\ \dfrac{{\rm d}N_2(t)}{{\rm d}t} = \lambda_1 N_1(t) - \lambda_2 N_2(t) - \sum\limits_j \sigma_2(E_j)\phi(E_j)N_2(t) \end{cases}, \tag{4}$$

Y is the cumulative yield of the mother nucleus from the nuclear fuel and λ_1, λ_2 are the decay constants of the mother and daughter nuclei, respectively. σ_1, σ_2 are the cross sections to form the mother and daughter nucleus via neutron capture reactions. In some of the decay chains, such as ^{90}Se$\longrightarrow$ Br $\longrightarrow$ Kr $\longrightarrow$ Rb $\longrightarrow$ Sr, the element at the end of the chain (^{90}Sr) has a long half-life (28.78a), while the half-life of the others is less than 3 minutes. In this case, the value of Y in Eq. (4) should be obtained by summing up the cumulative yields of all the short-half-life elements because of the radioactive equilibrium.

3.3 Generation from neutron capture

The equation is almost the same with Eq. (4):

$$\begin{cases} \dfrac{{\rm d}N_{\rm s}(t)}{{\rm d}t} = \sum\limits_i^{\rm isotopes} f_i(t)Y_i - \lambda_1 N_{\rm s}(t) - \sum\limits_j \sigma_1(E_j)\phi(E_j)N_{\rm s}(t) \\ \dfrac{{\rm d}N_2(t)}{{\rm d}t} = \sum\limits_k \sigma_1(E_k)\phi(E_k)N_{\rm s}(t) - \lambda_2 N_2(t) - \sum\limits_j \sigma_2(E_j)\phi(E_j)N_2(t) \end{cases}. \tag{5}$$

Take ^{90}Y as an example. It can be generated from ^{89}Y (n,γ)^{90}Y. In Eq. (5), Y is the sum of the cumulative yield of the chain that A=89. λ_1, λ_2 are the decay constants of ^{89}Y, ^{90}Y, respectively, and σ_1, σ_2 are the cross sections of the (n,γ) reaction for ^{89}Y, ^{90}Y.

With the above calculation, we can determine the activities of the long half-life isotopes at the moment when the SNF is taken out of the reactor. Then, these elements will decay with their own half-life (or the mother nucleus's half-life due to the radioactive equilibrium), and their activity at any time can be obtained. The growth of these long half-life isotopes in a reactor during its running can also be calculated.

4 Calculation of the antineutrino spectrum

According to Ref. [9], if the possible effect associated with the unique first forbidden decays (and

higher forbidden decays) is neglected, then the electron spectrum can be assumed to be the shape:

$$N(E_e) = k(E_0, Z)E_e p_e(E_0 - E_e)^2 F(Z+1, E_e), \quad (6)$$

where k is a normalization constant, E_e and p_e are the full electron energy and momentum, and $F(Z+1, E_e)$ is used to correct the Coulomb effect on the outgoing electron, Ref. [10] shows a simple relation for the $F(Z+1, E_e)$. A unique first forbidden decay mode exists in this calculation, so a shape factor $S_n(Z, E)$ should be multiplied. It is described [11] as:

$$S(E) = (W-1)^2 + (W_0 - W)^2, \quad (7)$$

where W_0 and W are the maximum and total β^- particle energy, taking $m_e c^2$ as their unit.

The antineutrino spectrum as a function of the antineutrino energy E_ν is obtained from Eq. (6) by substituting $E_\nu = E_0 - E_e$ in the above formula.

With the spectrum shape, combining with the calculated fission rates of ^{235}U, ^{238}U, ^{239}Pu, ^{240}Pu, ^{241}Pu and ^{242}Pu for a typical 1 GW PWR[1)]. The activities of the long half-life isotopes listed in Table 1 can be calculated, and the antineutrino spectra from these isotopes can be determined at any time. Fig. 1 shows an example of the calculated results, assuming that the reactor was shut down to change its fuel on the 330th day.

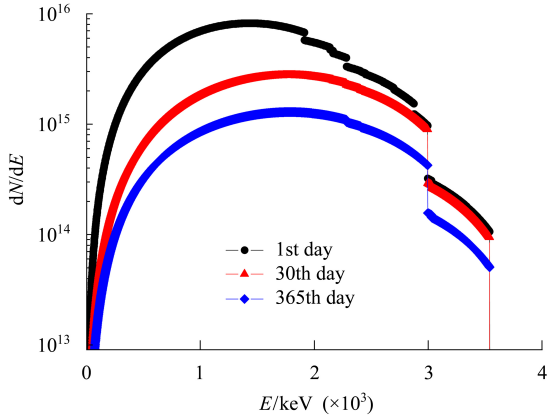

Fig. 1. The calculated antineutrino spectra from SNF after a typical 1 GW reactor is shut down for 1, 30 and 365 days, respectively.

5 The contribution of antineutrino spectra from long-life isotopes

Figure 2 shows the total antineutrino spectra converted from the measured beta spectra after continuous bombardment by neutrons on a nuclear fuel rod [12–14]. In these experiments, the measured beta spectra are from nuclear-fuel fission products with an exposure time to neutron of about 40 hours. Most of the isotopes from the fission could reach an equilibrium state straight after the bombardment, while as Fig. 3 shows, the total activity of the long-life isotopes increases when the reactor is running. So the contributions of the non-equilibrium state of the long-life isotopes are non-negligible, and the antineutrino spectra will be variational as the running time of the reactor. The antineutrino spectra obtained from the measured beta spectra that were acquired after a short irradiation time in a thermal neutron flux couldn't stand for the ones during the whole running time.

Due to this phenomenon, the following steps were performed to obtain the real total spectra (Fig. 4).

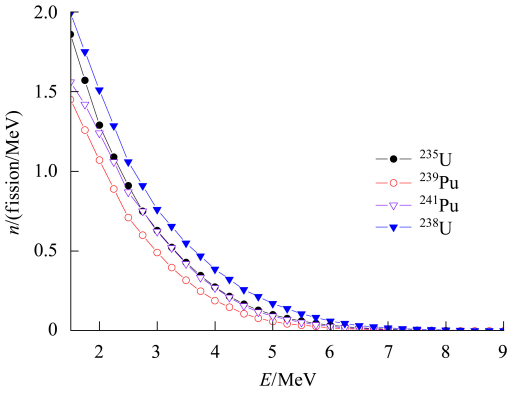

Fig. 2. Total antineutrino spectra converted from measured beta spectra after continuous bombardment by neutrons. For ^{235}U ^{239}Pu and ^{241}Pu, the exposure time to the neutron is 1.5, 1.8 and 1.8 days, respectively. As for ^{238}U, the spectrum was calculated with an infinite exposure time.

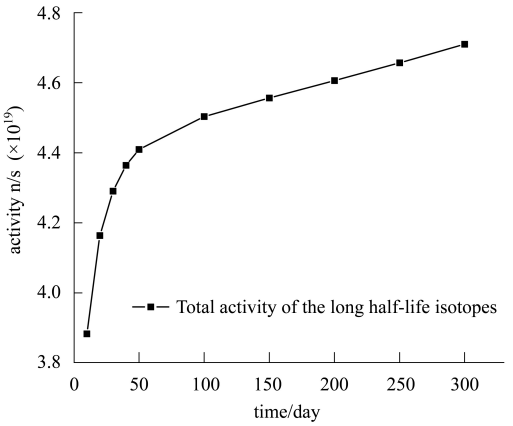

Fig. 3. Changes in the total activity of the long half-life isotopes during the running of the reactor. This is a state of non-equilibrium generation for the long half-life isotopes.

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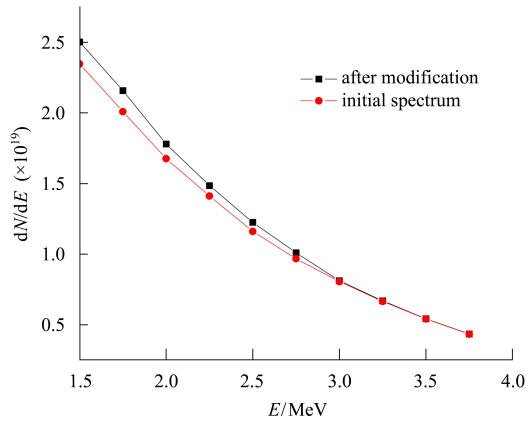

Fig. 4. Because of the non-equilibrium, as well as the maximal Q value of the long half-life isotopes (3.956 MeV), the spectrum from the 330th day, after correction, will be higher at the low-energy region compared with the original one.

1) The antineutrino spectra obtained from the measured beta spectra were corrected for the non-equilibrium part.

2) The total equilibrium antineutrino spectra were calculated with the equilibrium antineutrino spectra obtained from the above calculation and the fission rates of the fuel isotopes as a function of time.

3) The non-equilibrium antineutrino spectra were calculated.

4) The total antineutrino spectra will at any time be the sum of the spectra from the long half-life isotopes and the total equilibrium spectra at a certain moment (both are as a function of time).

With the above modification to the non-equilibrium part, the total spectrum of the 330th day can be calculated well. Fig. 5 illustrates, when the reactor is shut down, the ratio of the antineutrino spectra emitted from the long half-life isotopes in spent nuclear fuel, which was taken out from the reactor to the total spectra at the end of the operating cycle.

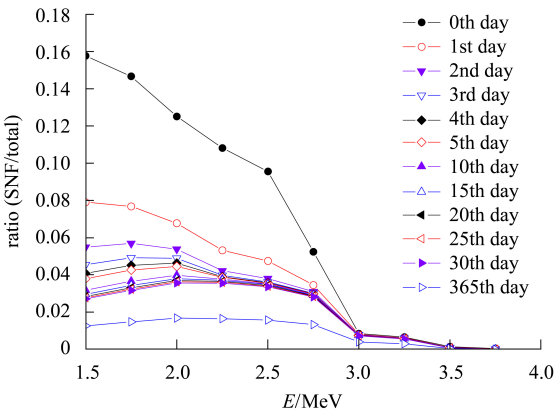

Fig. 5. The ratio of the antineutrino spectra from spent nuclear fuel to the total antineutrino spectrum at the end of the operating cycle. The numbers to the right of the arrow indicate the cooling time of the spent nuclear fuel.

6 The event rate

Combined with the cross section of the H($\overline{\nu}_e$, n)e^+ reaction, the ratio of the event rate from the long half-life isotopes to the total event rate, which was deduced from the total corrected antineutrino spectrum on the running days, can be calculated (Fig. 6).

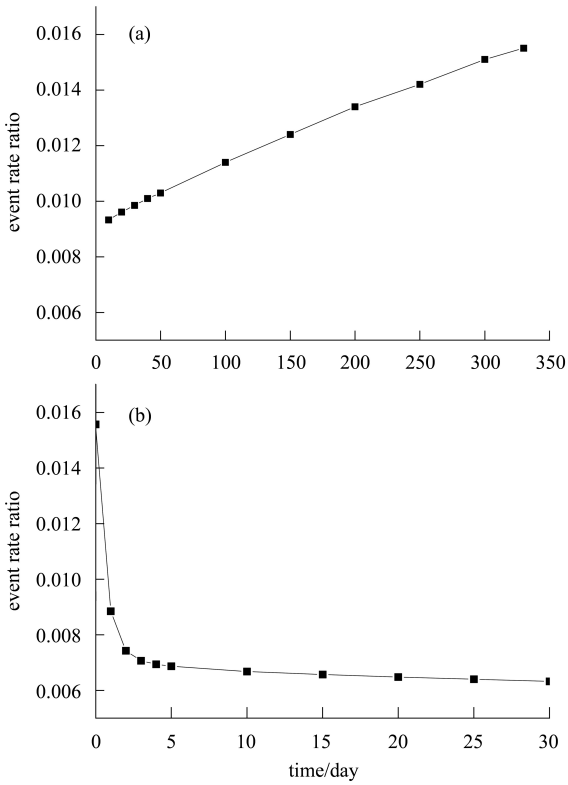

Fig. 6. The ratio of the events from the long half-life isotopes to the that from all the fission products when the reactor is on (a). The ratio of the events from spent nuclear fuel to the total events at the end of the operating cycle (b).

The results show that the event rate from the non-equilibrium antineutrinos from these long half-life elements contributes about 0.93–1.55 percent during the running of the reactor. And the event rate from the SNF is about 0.63% compared with the total event rate when the reactor is on, even after the SNF has been taken out of the reactor for more than 30 days.

Since the amount of SNF stored in the storage pool near the reactor core is much larger than the fuel in the core, the event rate and its impact on the experimental results from the SNF must be well estimated for such precise experiments.

7 Summary and discussion

In summary, taking into account the operation of a typical 1 GW reactor, a detailed calculation of the generation of long half-life isotopes in a nuclear reactor, and the antineutrino spectrum emitted from SNF, has been performed. The effect of the non-equilibrium of the reactor antineutrino spectrum has been shown.

This calculation was based on an ideal reactor operation cycle. In reality, a reactor needs to stop twice to change fuel during a full running cycle, but only about one-third of the fuel was changed each time. This will lead to the different results from this calculation. We estimate that in a real reactor, the event rate from the long half-life elements will contribute about 1.5%–2.0% during the running of the reactor. We also estimate that the event rate contribution from the SNF stored in the Daya Bay and Linao should be about 1.9%, considering the running history of these reactors (we assume that all SNFs are stored in the cooling pools at Daya Bay and Linao from the beginning). For the soft part, this contribution would be much larger, since the maximum antineutrino energy from SNF is below 4 MeV. The event rate contribution from SNF in the 1.8–4.0 MeV region would be larger than 4%.

This work developed a method to calculate antineutrinos from long half-life isotopes from a reactor. This is a non-equilibrium part during the running of the reactor, and is also a continuous "background" after the fuels are removed to SNF pools near the reactor cores. The influence and impact of these neutrinos on the experiment should be studied and incorporated into the data analysis, and combined with the detailed reactor running data provided by the nuclear power plant.

References

1 A Precision Measurement of the Neutrino Mixing Angle θ_{13} Using Reactor Antitrinos at Daya Bay. Daya Bay Proposal, December 1, 2006. arXiv: hep-ex/0701029v1, 2007

2 Kopeikin V I, Mikaelyan L A, Sinev V V. Phys. Atom. Nucl. 2001, **64**: 849–854 [arXiv:hep-ph/0110290v1]

3 Kopeikin V. Phys. Atom. Nucl. 2003, **66**: 472–475; Yad. Fiz. 2003, **66**: 500–503[arXiv:hep-ph/0110030]

4 Kopeikin V I, Mikaelyan L A, Sinev V. Phys. Atom. Nucl. 2006, **69**: 185–188 [arXiv:hep-ph/0412044v1]

5 Kopeikin V I, Mikaelyan L A. Phys. Atom. Nucl. 2006, **69**: 1888–1893 [arXiv:hep-ph/0508239v1]

6 AN Feng-Peng et al. Chinese Physics C (HEP & NP), 2009, **33**: 711–716

7 http://www.nndc.bnl.gov/exfor/endf02.jsp, target=^{235}U, ^{239}Pu, ^{240}Pu, ^{241}Pu, ^{242}Pu, Projectile=*fpy*, MT=459 http://www.nndc.bnl.gov/exfor/endf00.jsp, target=^{89}Y et al. Reaction=n,g. MT=102, LR=0

8 Hitoshi IHARA et al. JNDC FP Decay and Yield Data. JAERI-M. 1981.09

9 Vogel P. Phys. Rev. C, 2007, **76**: 025504

10 Brabec V, Rysavy M. Europhys. Lett., 1993, **21**(8): 811–816

11 LU Xi-Ting et al. Nuclear Physics. Beijing: Atomic Energy Press, 2000.10. 148 (in Chinese)

12 von Feilitzsch F, Hahn A A, Schreckenbach K. Phys. Lett. B, 1982, **118**: 162–166

13 Hahn A A et al. Phys. Lett. B, 1989, **218**: 365–368

14 Vogel P et al. Phys. Rev. C, 1981, **24**: 1543–1553