The energy calibration system for CANDLES using (n,γ) reaction

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Abstract

CAlcium fluoride for the study of Neutrinos and Dark matters by Low-energy Spectrometer (CANDLES) is searching for neutrino-less double-beta decay of ^{48}Ca using a CaF_2 scintillator array. A high Q-value of ^{48}Ca at 4,268 keV enabled us to achieve low background, however, at the same time it causes difficulties in calibrating the detector's Q-value region because of the absence of a standard high-energy γ-ray source. Therefore, we have developed a novel calibration system based on γ-ray emission by neutron capture on ^{28}Si, ^{56}Fe, and ^{58}Ni nuclei. In this paper, we report the development of the new calibration system as well as the results of energy calibration in CANDLES up to 9 MeV.

Keywords: Double beta decay, ^{48}Ca, Energy calibration, (n,γ) reaction, Scintillation detector, Calcium fluoride

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1. Introduction

Because the discovery of neutrino oscillations has revealed the existence of a neutrino mass [1, 2], the search for a neutrino-less double-beta decay ($0\nu\beta\beta$) has become one of the preeminent topics in modern physics. Double beta decay, which emits two neutrinos ($2\nu\beta\beta$), has been observed by several experiments[3]. However, $0\nu\beta\beta$ has remained unobserved till date. If decay would be observed, this would imply that the neutrino is a Majorana particle, and that the lepton number conservation is violated. Accordingly, the asymmetry of matter and antimatter in the present universe can theoretically be explained by the leptogenesis scenario [4]. As a Majorana particle, the neutrino emitted by a neutron via beta decay can be absorbed by another neutron in the same nucleus. The $0\nu\beta\beta$ thus produces a mono-energetic signature at the end of the $2\nu\beta\beta$ energy spectrum. The rate of decay increases alongside the square of the effective neutrino mass; therefore, its measurement can provide information on absolute neutrino mass scale. Since $0\nu\beta\beta$ is expected to be an extremely rare signal, and its half-life is longer than 10^{26} year [5, 6], low radioactive contamination and good energy resolution is required to separate a mono-energetic signal from background noise. Two-neutrino mode of double-beta decay can also be a serious source of background due to non-perfect energy resolution.

CAlcium fluoride for the study of Neutrinos and Dark matters by Low energy Spectrometer (CANDLES) is a ^{48}Ca double beta decay experiment using CaF_2 scintillator and photomultiplier tubes (PMT) [7]. The CANDLES-III detector was constructed at the Kamioka Underground Observatory in Japan and is currently collecting experimental data [8, 9]. The Q-value of ^{48}Ca (i.e., the sum of the kinetic energies of the two electrons emitted at $0\nu\beta\beta$) is 4,268 keV [10]. It is the largest among the candidate nuclei for double beta decay. Taking advantage of this, CANDLES aims to measure $0\nu\beta\beta$ in an ultra-low background environment [11]. The high Q-value of ^{48}Ca makes it difficult, however, to precisely calibrate the detector due to the absence of a high-energy standard gamma ray source. To date, we have calibrated the detector using a 1,836 keV γ-ray obtained from a ^{88}Y source. This is relatively high energy among commercially available radioactive γ-ray sources. In addition, we also employed a 2,615-keV γ-ray from ^{208}Tl, the source of which was radioactive contamination in the detector components. A higher energy calibration source is required because precise energy calibration near the Q-value region is very important for the identification of $0\nu\beta\beta$ peak. Herein, we report the construction of an energy calibration system above 3 MeV using gamma rays from neutron capture reactions on Si, Fe, and Ni. Moreover, we report the energy calibration of the CANDLES detector using the above system.

2. Detector Overview

The CANDLES-III detector (Figure 1) operates with 305-kg CaF_2 crystals in the Kamioka Underground Observatory. The detector comprises 96 pure calcium fluoride (CaF_2) crystals sized 10^3 cm^3 immersed in a liquid scintillator (LS) as a 4π active shield. The crystals were divided into six layers in a vertical (Z) direction, and each layer included 16 crystals. The LS has a much shorter decay time constant ($\sim$10 ns) compared with CaF_2 ($\sim$1 μs). Events of multiple energy deposition could thus be rejected by pulse shape analysis. Scintillation light from CaF_2 and the LS is observed by 62 PMTs with light-collecting pipes mounted on a 30 m^3 stainless steel water tank. Since CaF_2 crystals are generally used in camera lenses and other commercial applications, comparative crystals with high transparency and high purity have been developed and are available. Because of the long attenuation length of CaF_2 and the LS ($>$ 10 meters), the scintillation light was collected by PMTs without attenuation, and thereby accurate energy information was obtained. The wavelength of the scintillation light of the CaF_2 crystals was shifted from 280 to 420 nm by a wavelength shifter (Figure 1) to effectively collect photons with PMTs [12]. The CANDLES experiment in this work adopted a data acquisition (DAQ) system using the SpaceWire protocol and the DAQ-Middleware framework. Because of the long decay time constant of CaF_2, a dual gate trigger logic was developed. Following the trigger, 8-bit 500 MHz flash analog-to-digital converters (FADCs) opened a 8,960-ns window and read out all PMT waveforms. Details of the CANDLES DAQ system are summarized in [13, 14].

The room where the experiment was conducted was cooled to approximately 2 °C. The detector temperature was maintained at 4 °C with a stability of 0.2 °C. The light yield of the CaF_2 scintillator is known to vary with temperature. If the temperature is lowered by 1 °C, the light yield increase by roughly 2 %. High-purity crystals can achieve very low levels of radioactive impurities, resulting in very low levels of radioactive background. Radioactive impurity concentration for each crystal was distributed between a few to a few tens [μBq/kg] in the Th chain.

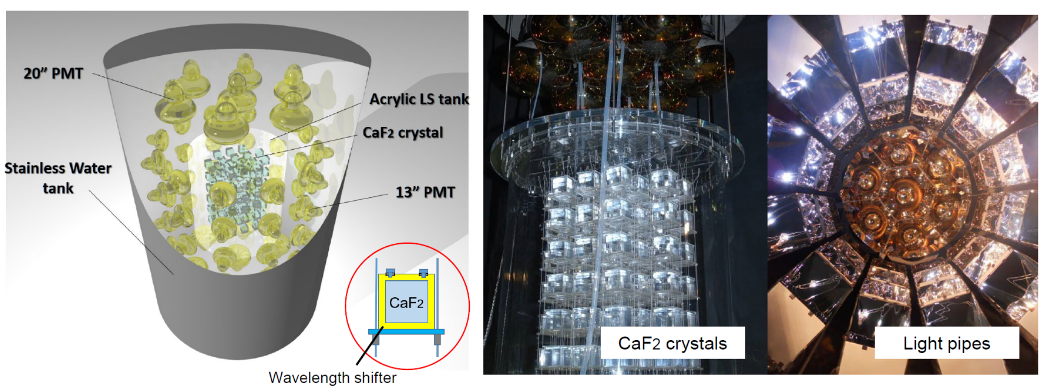

Figure 1: The figure on the left presents a schematic view of the CANDLES-III detector. The stainless steel tank has a 3-m diameter × and a 4-m height. The acrylic tank is 1.4 m in diameter × and has a height of 1.4 m. The size of the CaF_2 crystal is 10^3 cm^3 cubic. Light pipes have been omitted from the figure for better visibility. Two images on the right show the CANDLES CaF_2 crystal array and light pipes as seen from above the tank.

3. Current Calibration in CANDLES

Energy calibration is important in double beta decay experiments because it determines the region where signals are expected but nothing appears usually. We calibrated each crystal with the aim of achieving 0.5% precision that is adequately smaller than the energy resolution at the Q-value region. As the radioactive backgrounds from internal and external sources are very low, the statistical significance of the data from these backgrounds is very poor. Accordingly, we placed a calibration source of ^{88}Y , with γ-rays at 898 and 1,836 keV, in the tank for relative gain calibration between crystals, with sufficient statistical accuracy. The commercially available ^{88}Y source had maximum activity of 50 kBq, and was placed in the liquid scintillator tank inside the crystal array. Events in each crystal were selected via position reconstruction, and CaF_2 crystal events were selected by pulse shape analysis, which removed LS energy deposition larger than approximately 100 keV at 1,836 keV. Based on the above procedure, we successfully performed energy calibration for each crystal using a γ-ray energy peak at 1,836 keV within 0.3 % statistical precision.

After relative calibration by the ^{88}Y source, we corrected the absolute energy scale using an external γ-ray from ^{208}Tl, the highest energy of which was 2,615 keV closer to the Q-value of ^{48}Ca. ^{208}Tl is a daughter of ^{232}Th presented as impurity in the set-up and environmental materials. Thus, the event rate was not high enough to affect crystal-by-crystal calibration with good precision. Since we found a small % layer dependence of energy scale in the 2,615-keV γ-ray, even after ^{88}Y calibration, energy scale correction was applied for each layer (i.e., 16 crystals).

4. New Calibration Method

Since Q-value was significantly higher than the current calibration point at 2,615 keV, we had to check the energy scale linearity at 4,268 keV. We developed a high-energy γ-ray calibration source by means of (n,γ) reaction. In general, several MeV γ-rays were emitted by neutron capture reaction on various nuclei. The ^{28}Si (n,γ) reaction was to some extent unique. The excited state of ^{29}Si following neutron capture emitted 3,539-keV γ-ray by decaying to an excited state at 4,934 keV, which in turn emitted 4,934-keV γ-ray [15, 16]. Since the energies of these γ-rays were close to the Q-value, they were able to provide a reasonable energy calibration for CANDLES. However, the small cross section of 0.17 barn for thermal neutron capture reaction required an effective neutron moderation system. This will be described later in this paper. Other neutron-capture γ-rays used in this system are summarized in table 1. The branching ratio was calculated as the ratio of the cross section of each branch to the total cross section of the isotope, which was taken from the database of International Atomic Energy Agency (IAEA) [17]. With the above γ-rays, the energy scale at Q-value (4,268 keV) could be obtained by interpolation. To date, however, we have relied on extrapolation (see Figure 2).

In this chapter, we describe the development of a new calibration system using the (n,γ) reaction of ^{28}Si, ^{56}Fe, and ^{58}Ni to affect calibration in the energy region of 3 MeV and higher.

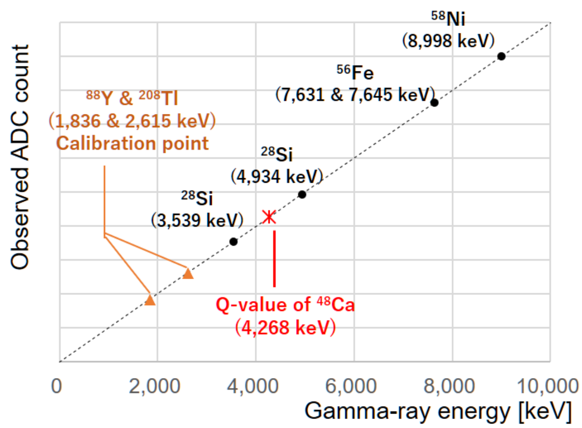

Figure 2: Conceptual plot for the CANDLES energy calibration. The cross marker shows the Q-value of ^{48}Ca; the triangle indicates the current calibration points at 1,836 keV for ^{88}Y and 2,615 keV for ^{208}Tl. The black circles show the new calibration point added in the present study.

Table 1: The γ-rays from the thermal neutron capture used in this calibration system. The neutron capture cross section emitted γ-ray energy and a γ decay branching ratio from an excited state (refer to database [17].)

	Neutron cross section	γ-ray energy	Branching ratio
^{28}Si	0.177 barn	3,539 keV	67.2 %
		4,934 keV	63.3 %
^{56}Fe	2.59 barn	7,631 keV	25.2 %
		7,645 keV	21.2 %
^{58}Ni	4.50 barn	8,998 keV	33.1 %

4.1. Development of Silicon and Nickel blocks

Considering expandability and portability, we developed polyethylene blocks including metallic silicon (Si) and nickel oxide (NiO), referred to as a Si or Ni block, respectively. Each block was 20 cm × 10 cm × 5 cm. An image of a Si block is shown at the bottom right in Figure 3. Powdered Si or NiO was mixed with polyethylene and solidified using an epoxy adhesive. Since polyethylene and epoxy adhesive include large amounts of hydrogen atoms, fast neutrons are efficiently thermalized and captured on Si or Ni nuclei inside blocks. The composition of Si and Ni blocks are summarized in Table 2. In total, thirty-six and eight blocks were made for Si and Ni, respectively, to obtain a sufficient amount of (n,γ) events under CANDLES detection efficiency.

Table 2: Composition of Silicon and Nickel blocks

	Component material	Weight ratio
Si block	Metallic silicon (Si)	63.5 %
	Polyethylene	16.5 %
	Epoxy adhesive	20.0 %
Ni block	Nickel oxide (NiO)	22.5 %
	Polyethylene	42.5 %
	Epoxy adhesive	35.0 %

4.2. Calibration Setup on CANDLES

Calibration data were taken with the blocks set on top of the CANDLES detector (see Figure 3). The ^{252}Cf neutron source, the decay rate of which was roughly 200 kBq, was placed at the center of the blocks. The decay of ^{252}Cf included 3% spontaneous fission and emitted an average of 3.8 neutrons with a mean energy of approximately 2 MeV. Fast neutrons were moderated by multiple scattering on hydrogen atoms inside the blocks or in the surrounding paraffin blocks. The γ-ray emitted by moderated neutron capture on Si or Ni nuclei was detected by the CANDLES detector and used for detector calibration. Since ^{252}Cf emitted not only neutrons but also many γ-rays, a 5-cm Pb shield was placed in the CANDLES direction in order to reduce such background γ-rays.

We conducted three types of calibration runs with different setups, as well as a background run. The Si run used 36 Si blocks and the Ni run employed eight Ni blocks; blocks corresponded to 34 kg Si and 2.3 kg Ni, respectively. In the ^{252}Cf run, the ^{252}Cf source was placed on the stainless steel tank directly in order to generate the γ-ray from the ^{56}Fe neutron capture contained in the stainless steel tank. The conditions of three calibration runs and a background run are summarized in table 3. Total calibration data collection period of approximately 3 days is determined to ahieve the required statistical accuracy within 0.5 % for each γ-ray peak analysis.

Insertion of a neutron source in a low-background experiment may cause an increase in the background rate due to activation of the detector material by neutron capture. However, due to the high Q-value of ^{48}Ca, long-lived isotopes that emit high-energy γ-ray above Q-value are not produced by neutron capture. Consequently, we reasoned that the background rate at Q-value was not affected by the use of the neutron source.

5. Data Analysis

Detailed analysis of the obtained data was carried out offline. Figure 4 shows the energy spectra in three different setups. X-axis is a observed energy

Table 3: Summary of the Calibration Runs for Each Setup

Run	Setup	Duration
Si run	Si block (34 kg of Si)	50.7 hours
Ni run	Ni block (2.3 kg of Ni)	12.1 hours
^{252}Cf run	^{252}Cf on tank	12.0 hours
Background run	No source	131 days

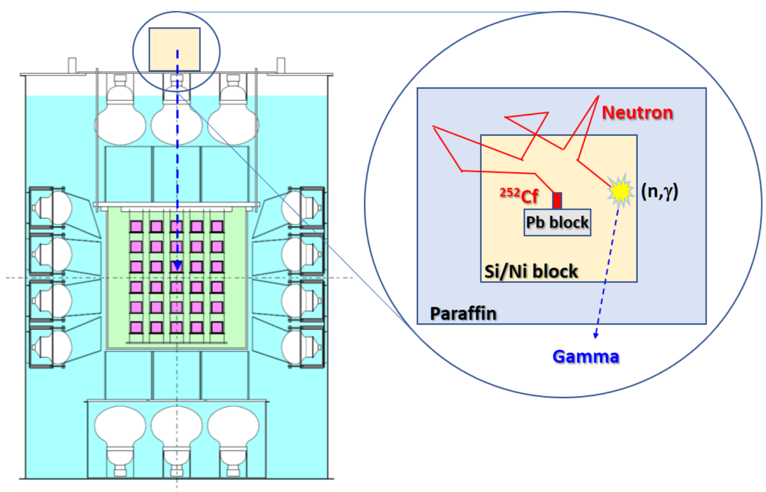

Figure 3: Schematic view of the calibration system. Si and Ni blocks were set on top of the CANDLES detector. Neutrons emitted from ^{252}Cf were moderated by collision on hydrogen atoms and finally captured by ^{28}Si or ^{58}Ni nuclei, followed by γ-ray emission.

calibrated by ^{88}Y and ^{208}Tl. The pulse shape analysis was performed event by event, and the events that included the light emission of LS were removed (LS cut). The LS cut removed more than 99 % of events with LS energy deposition of 100 keV at the visible energy of 1,836 keV. This was applied to observe clear peaks through a reduction of Compton scattering events. The γ-rays' energy peaks from ^{28}Si, ^{56}Fe, and ^{58}Ni shown in table 1 were apparent in the data of the Si, ^{252}Cf, and Ni runs, respectively.

A 6 MeV peak was coming from 5,920-keV and 6,018-keV γ-rays of neutron capture of ^{58}Fe nuclei. Peak around 6.8 MeV in Ni run data was combination of 6,584-keV, 6,720-keV, and 6,838-keV γ-rays from neutron capture of ^{58}Ni, ^{60}Ni, and ^{62}Ni, respectively. These γ-rays are not used in our analysis because large peak-energy difference affects the energy resolution analysis adversly. Moreover, peak of 2,223 keV is neutron capture γ-ray of hydrogen inside the water, which is also apparent only in the neutron source calibration data.

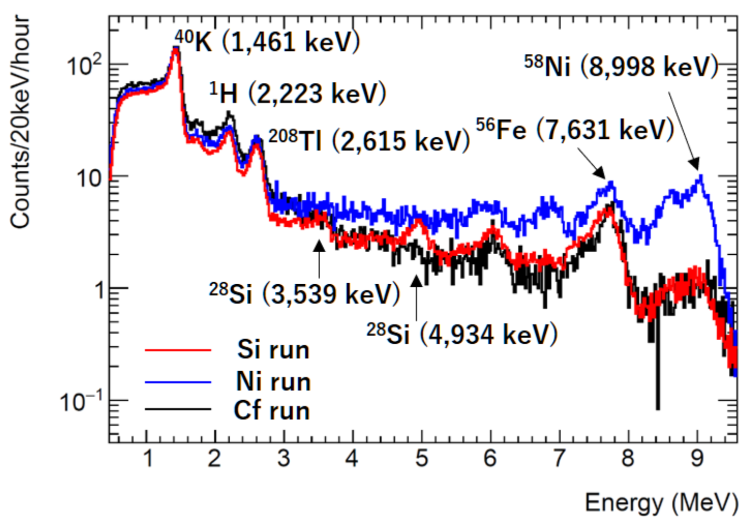

Figure 4: Energy spectra for Si run (red), Ni run (blue), and ^{252}Cf run (black) following the LS cut normalized by the live time of each run. Characteristic peaks appear in each run.

In order to extract a signal from the obtained spectrum, fitting was performed under the assumption that the signal and background components comprised Gaussian, exponential, polynomial, and constant elements. Prior to this analysis, the response of the CANDLES detector to γ-rays and the validity of the above background assumption were confirmed in a study based on Monte Carlo simulation and real data.

Figure 5 show the fitting results of each γ-ray energy peak. The two upper images show ^{58}Ni and ^{56}Fe; the bottom left represents 4,934 keV of ^{28}Si, and the bottom right that of 3,539 keV of ^{28}Si. In the case of Ni, an exponential, a constant, and a Gaussian element were assumed as background for the 8,533 keV γ-ray of ^{58}Ni in order to produce the 8,998-keV peak of ^{58}Ni. In this figure, 8,533 keV of ^{58}Ni is visible; however, it was not used for calibration because this energy region may have been affected by the Compton edge of the 8,998-keV γ-ray. The ^{56}Fe emitted two very close energy γ-rays with energies of 7,631 and 7,645 keV and branching ratios of 25.2 % and 21.2 %, respectively. Although data included these two γ-rays, their energy difference was much smaller than the energy resolution of CANDLES ($\sigma \sim 140$ @ 7,631 keV). These two peaks were thus treated as one Gaussian distribution with a mean of 7,637 keV, the weighted average of two γ-rays taking into account the emission probability of each γ-rays. In addition, the 7,279 keV γ-ray of ^{56}Fe and a constant component were assumed as background during fitting. In the analysis of the energy spectrum in the energy regions of 4,934-keV and 3,539-keV peaks, the background was not clearly known. The shape of the background was thus assumed by an appropriate combination of Gaussian, exponential, polynomial, and constant elements. We attempted several different combinations and confirmed that the results varied within a statistical error at most. Due to the relatively small cross section of ^{28}Si, it was anticipated that contamination by higher energy γ-ray events (which partially deposited energy into CaF_2 by Compton scattering) could render analysis difficult. However, the obtained results from ^{28}Si analyses were reasonable. The statistical error in the Si peak analysis is about 0.2 % for 4,934 keV and 0.3 % for 3,539 keV, and it is considered that the enough statistics to ensure the linearity have been obtained.

The energy linearity and energy resolution of CANDLES were studied using

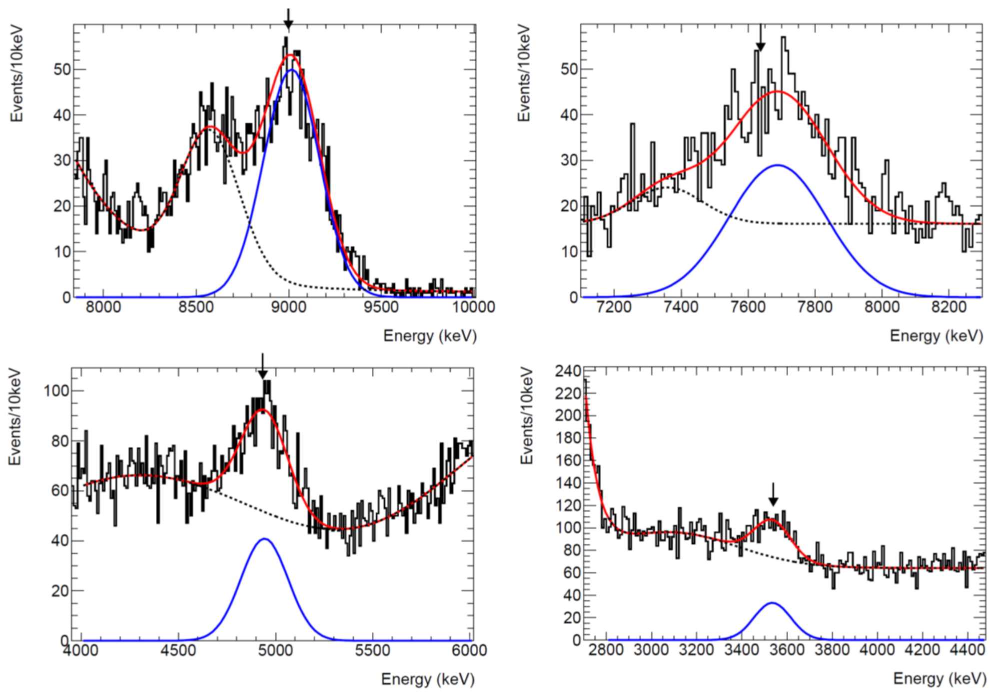

Figure 5: Fitting results for 8,998 keV of ^{58}Ni (top left), 7,631 + 7,645 keV of ^{56}Fe (top right), 4,934 keV of ^{28}Si (bottom left), and 3,539 keV of ^{28}Si (bottom right), respectively. The red line shows the fitting results, the black dashed line shows the background, and the blue line represents the signal. The arrows are corresponding to the prospective energy of γ-ray.

the results of each peak fitting. Figure 6 shows the linearity of energy response as a function of observed γ-ray energy. The Y-axis of the bottom plot is a ratio of observed energy and prospective energy for each γ-ray. Observed energy is peak energy extracted by fitting. Prospective energy refers to the database of the International Atomic Energy Agency [17]. Four additional calibration points above 3 MeV became available following installation of the new calibration system and the energy scale at the Q-value region was obtained by interpolation. From this figure, good energy linearity for CANDLES up to 9 MeV was confirmed and energy deviation at the Q-value region was estimated to be less than 0.3 %. Until now, the energy scale was calibrated at 2,615 keV, and the linearity at higher energies could not be confirmed. With the calibration by 2,615-keV γ-ray of ^{208}Tl, deviation of linearity is about 0.9 % at 1,836 keV. If this is scaled by the Q-value of 4,268 keV, it becomes an uncertainty of approximately 2 %. For the first time, with the present calibration, it was confirmed that the energy scale at the Q-value was linear within 0.3 %. This is very important result for our $0\nu\beta\beta$ search in CANDLES.

The nonlinearity of the scintillation efficiency with respect to the γ-ray energy in the lower energy region below 3 MeV confirmed by the series of calibration is considered to be a characteristic of the pure CaF_2 scintillator. This kind of non-linearity commonly appears in inorganic scintillator detector. Inside the scintillator, kinetic electrons are generated by Compton scattering of incident γ-ray, and the energy of this electron depends on the energy of the original γ-rays. The energy loss (dE/dx) of the electron depends on the energy, thereby it is thought that such non-linearity appears [18, 19].

Figure 7 shows the energy resolution for CANDLES as a function of observed energy. Each circle point corresponded to γ-ray measured in the calibration or background run. The Y-axis of the plot was a fitted σ for each γ-ray peak. In the resolution analysis, 2,223 keV γ-ray from hydrogen neutron capture is also included, as shown in the figure. However, ^{40}K was not used for the fitting this time, because, due to large statistics, it requires the detailed study for an appropriate fit function such as contamination from the low energy side by

Compton scattering.

Simple single exponential fitting resulted in roughly $\sigma = 2.4 \pm 0.2$ % at 4,268 keV. The energy resolution that is very important for $0\nu\beta\beta$ search was also obtained for the first time above 3 MeV by introducing the new calibration system.

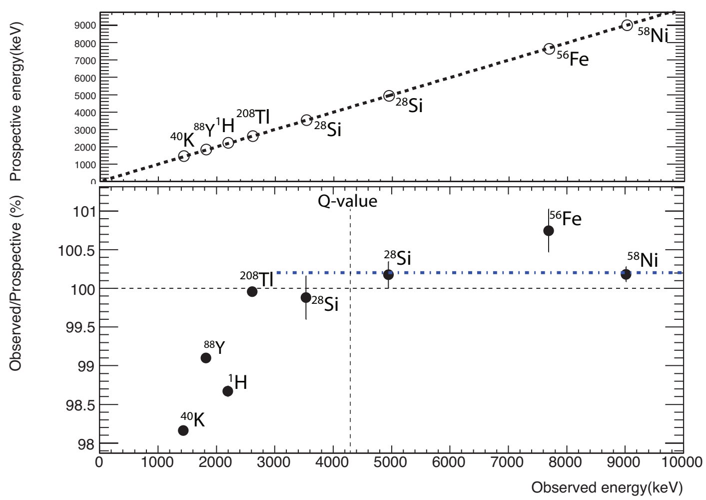

Figure 6: The top image shows prospective energy vs observed energy. Each point corresponds to γ-ray peak energy measured by calibration or background run. The bottom image shows energy deviation for CANDLES as a function of observed energy. Dash-dot line is an average obtained from four new calibration points. Good linearity of energy response within 0.3 % was obtained in the region of interest by this work. Error bar shows only statistical error.

6. Conclusion and Outlook

In this paper, we reported a new energy calibration method for CANDLES to search for neutrino-less double beta decay of ^{48}Ca. A high Q-value of ^{48}Ca may be a key to performing very low background $0\nu\beta\beta$ measurements. To date, an absolute energy scale of CANDLES was calibrated at 2,615 keV, despite a high Q-value of 4,268 keV. We designed and developed the calibration method using γ-rays from (n,γ) reactions. This enabled us to calibrate energy up to 9

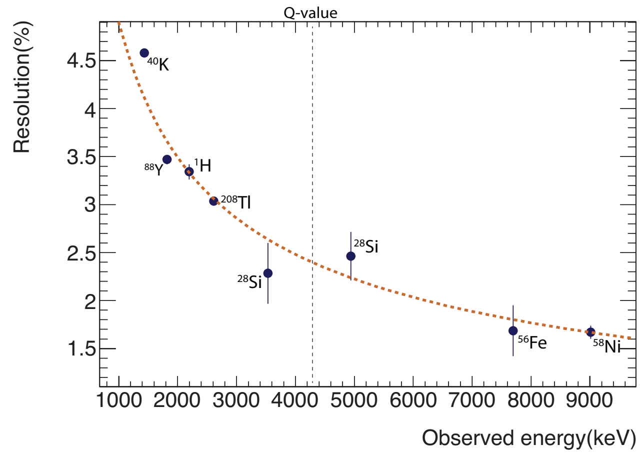

Figure 7: Energy resolution (σ) for CANDLES as a function of observed energy. The circle marker indicates γ-ray observed in CANDLES by calibration and background run. Single exponential function indicated good fit with the data up to 9 MeV (dashed line). The error bar indicates only statistical error.

MeV.

Four calibration points at 3,539 keV, 4,934 keV of ^{28}Si, 7,631 + 7,645 keV of ^{56}Fe, and 8,998 keV of ^{58}Ni were realized by introducing the present energy calibration system. The obtained results provide confirmation of good energy linearity above 3 MeV for the first time in a CANDLES experiment with uncertainty below 0.3 % at 4,268 keV, which is adequately smaller than the resolution. Additionally, energy resolution at the Q-value was measured and estimated to be 2.4 $\pm$ 0.2 % at 4,268 keV.

In future research, the background must be reduced in order to see clear peaks and to limit the number of statistical errors. Peaks of ^{28}Si were contaminated by the 7,631-keV γ-ray of ^{56}Fe, which partially deposited the energy by Compton scattering in CaF_2. An additional neutron shield on the stainless steel tank in the Si run should effectively reduce such background noise and ensure better energy calibration for the CANDLES experiment.

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