### **ARTICLE**

## Upgrading of shielding for rare decay search in CANDLES

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In the CANDLES experiment aiming to search for the very rare neutrino-less double beta decays  $(0\nu\beta\beta)$  using  $^{48}$ Ca, we introduced a new shielding system for high energy  $\gamma$ -rays from neutron captures in massive materials near the detector, in addition to the background reduction for  $^{232}$ Th decays in the  $0\nu\beta\beta$  target of CaF<sub>2</sub> crystals. The method of background reduction and the performance of newly installed shielding system are described.

Keywords: double beta decay; underground experiment; low background measurement;  $\gamma$ -rays from neutron captures

### 1. Introduction

The neutrino-less double beta decay  $(0\nu\beta\beta)$  is a process beyond the standard model and has not been observed yet. The existence of  $0\nu\beta\beta$  leads to three important facts; (1) The lepton number violation, which is a key of our matter dominated universe. (2) The Majorana nature of neutrinos, which indicates that particle is its own anti-particle. (3) The absolute scale of neutrino mass. A mass hierarchy of neutrinos is determined by the  $0\nu\beta\beta$  observation.

In  $0\nu\beta\beta$ , two electrons are emitted and the  $0\nu\beta\beta$  measurement tries to find the signals at sum energies of electrons corresponding to the Q value. The  $0\nu\beta\beta$  is a quite rare phenomenon and low background techniques are required to observe such rare decays. Until now, only a lower limit of half-life was experimentally obtained to  $10^{26}$  year by the KamLAND experiment using  $^{136}$ Xe [1].

CANDLES (CAlcium fluoride for the study of Neutrinos and Dark matters by Low Energy Spectrometer) is a double beta decay experiment using <sup>48</sup>Ca in CaF<sub>2</sub> crystals [2]. The advantage of <sup>48</sup>Ca is the highest Q value among all isotope candidates (4.3 MeV), and therefore the background sources are limited in such

a high energy region. The disadvantage of  $^{48}$ Ca is a low natural abundance of  $\sim 0.2\%$ , and the enrichment technique is under development [3].

The best result thus far obtained for a lower limit of half-life using  $^{48}\text{Ca}$  is  $5.8\times10^{22}$  year (90% Confidence Level (C.L.)) by the ELEGANT VI experiment [4]. It was found that reduction of  $\gamma\text{-ray}$  background due to neutron capture around the detector is indispensable to improve this limit (section 3). This paper describes establishing a new shield system for the purpose.

#### 2. Detector

The CANDLES detector is located in the Kamioka underground laboratory (Gifu Pref., Japan) where the overburden is 2,700 m.w.e. (meter water equivalent). The muon flux is about 10<sup>-5</sup> of the ground level, and it leads to the lower muon-related environmental radiations such as neutrons.

A schematic view of the detector is shown in **Figure 1**. We use 96 CaF<sub>2</sub> crystals ( $10^3$  cm<sup>3</sup>) which contain 305 g of <sup>48</sup>Ca, as the scintillator detector and the  $0\nu\beta\beta$  target. The scintillation light from CaF<sub>2</sub> crystals are observed by 62 photomultiplier tubes (PMTs) with the light pipe system which has a 93% reflectivity. The CaF<sub>2</sub> crystals are immersed in the liquid scintillator (LS) which works as active veto, and an acrylic tank containing LS is set in

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the water passive shield with an average thickness of  $\sim$ 1 m. The whole system is installed in a stainless steel tank with 3 m diameter and 4 m height.

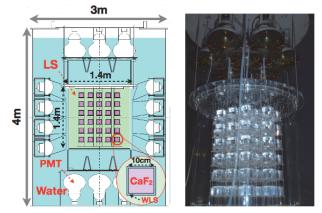


Figure 1. A schematic view of the detector. Left: A drawing of the detector from the side view. The whole detector is installed in a cylindrical stainless steel tank. Right: A photo of  $96\ CaF_2$  crystals which are immersed in LS.

A distinction of the experiment is a  $4\pi$  active shield by LS. The time constant of the pulse shapes are  $\sim 1~\mu$  sec and  $\sim 10$  nsec for CaF<sub>2</sub> and LS, respectively. When external  $\gamma$ -rays react with LS, the observed pulse shapes are different from that of the signal (i.e. the pulse of  $\beta$  rays in CaF<sub>2</sub>). By using this pulse shape difference, events which contain the LS pulse are rejected event by event by analyzing the pulse-shape data which are recorded by a 500 MHz FADC (Flash ADC) (LS cut). This active system is also useful for rejection of internal  $\beta$  decays in CaF<sub>2</sub> when emitted  $\gamma$ -rays deposit their energy in LS.

## 3. Background

#### 3.1. Background candidates

The background sources are suppressed thanks to the high Q value of  $^{48}\text{Ca}$ . There are three background candidates around 4.3 MeV. As shown in **Figure 2**, two of them are decay chain of a radioactive impurity,  $^{232}\text{Th}$  in CaF<sub>2</sub> crystals; (1)  $^{208}\text{Tl}$  ( $\beta$  decay,  $Q_{\beta}=5.0$  MeV), (2) Pile-up events of  $^{212}\text{Bi}$  and  $^{212}\text{Po}$  (Maximum energy is 5.3 MeV) due to short half-life of  $^{212}\text{Po}$  ( $\sim\!300$  ns). Since the time window of the pulse shape is 4  $\mu$  sec, the signals due to  $^{212}\text{Bi}$  and  $^{212}\text{Po}$  decays are observed as a single pulse.

The third background is high energy  $\gamma$ -rays from neutron captures in massive materials near the detector, called as  $(n,\gamma)$  background. Despite of the measurement in low neutron flux and with the combination of the LS active shield and the water shield, the  $(n,\gamma)$  events proved the largest background as mentioned in section 3.3

# 3.2. 208Tl and 212Bi-212Po backgrounds

We employed pure CaF<sub>2</sub> crystals developed by choosing material powders which has a less radioactivity

before growing the crystals [2,3]. The average  $^{232}$ Th activity of 96 crystals is  $\sim$ 29  $\mu$ Bq/kg and 27 of them have an activity below 10  $\mu$ Bq/kg.

The backgrounds related to  $^{232}$ Th-decay series can be removed through the off-line data analysis (analysis cut) [4]. Since the decay of  $^{212}$ Bi ( $\beta$  decay) and  $^{212}$ Po ( $\alpha$  decay) are observed as a single event due to the short half-life of  $^{212}$ Po (300 ns), such events are rejected if time differences between  $^{212}$ Bi and  $^{212}$ Po are longer than 20 nsec, which is a limit to distinguish the double pulse. If time differences are shorter than 20 nsec,  $\sim$ 95% events can be rejected by using the pulse shape difference between  $\alpha$  particles and  $\beta$  particles in CaF<sub>2</sub> [5]. The total rejection efficiency of this "Bi-Po cut" for  $^{212}$ Bi- $^{212}$ Po background is about 99%.

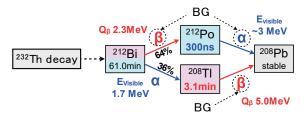


Figure 2. A schematic diagram of the <sup>208</sup>Tl and <sup>212</sup>Bi-<sup>212</sup>Po backgrounds in <sup>232</sup>Th decay chain.

To reject the  $^{208}$ Tl background, we apply veto time to the crystal where  $\alpha$  particles from a parent nuclei  $^{212}$ Bi are observed at 1.7 MeV (Figure 2). The veto time window is 12 minutes which is long enough compared to the  $^{208}$ Tl half-life, 3.1 minutes to inhibit collection of event due to the decay of  $^{208}$ Tl. The rejection efficiency of this " $^{208}$ Tl cut" is evaluated to be  $\sim 90\%$ . In order to achieve better rejection efficiency, it is necessary to apply veto time also for crystals neighboring to that where the  $\alpha$ -particles are observed although it causes larger dead time of the measurement time. It is because the position of the crystal is sometimes misidentified due to the interaction of successive  $\gamma$ -rays from daughter nucleus  $^{208}$ Tl with neighboring crystals. We call this cut as "multi crystal veto".

#### 3.3. $(n,\gamma)$ background

Even above Q value, nonnegligible amounts of events were observed. **Figure 3** shows the observed energy spectrum in the year-2013 data. Many events above Q value were observed even in the energy region from 7 to 8 MeV. The cause of these events was identified as high energy  $\gamma$ -rays from neutron captures in massive material around the detector such as rock and stainless steel. For example, the specific  $\gamma$ -ray energy from neutron captures on  $^{56}$ Fe is 7.6 MeV. These  $\gamma$ -rays sometimes deposit almost entire energy in CaF2 crystals and then remain as a background.

In order to confirm the origin of this background, we set a <sup>252</sup>Cf source outside the stainless steel tank to increase the neutron flux. The observed spectrum with a <sup>252</sup>Cf source is consistent with the data of normal run in the high energy region. The energy spectra with a

neutron source and in normal run were simulated by the Monte Carlo (MC) simulation using the code Geant4 [6]. As a result, the observed spectrum was well reproduced by considering  $\gamma$ -rays from neutron captures in rock and stainless steel. The  $(n, \gamma)$  background was estimated to be  $76\pm9(\text{statistical.})$  events/year/96crystals, which is beyond acceptable level.

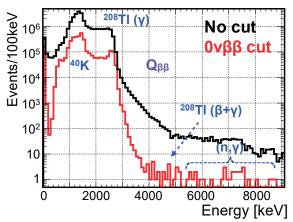


Figure 3. The observed energy spectrum in the year-2013 data. The black line shows the spectrum without analysis cuts, and the red line shows the one with all analysis cuts (0v $\beta\beta$  cut). We can see some events above Q $_{\beta\beta}$  (4.3 MeV). These events were identified as (n, $\gamma$ ) background.

### 4. Shield for (n,γ) background

## 4.1. Shield design

Since the  $(n,\gamma)$  events proved to be the most serious background in CANDLES, we installed an additional shield in the detector. The goal of the  $(n,\gamma)$  background rate is about 1 events/year/96crystals, which is  $\sim 1/80$  of the event rate in **Figure 3**. A shield design was optimized by Geant4 simulation. A schematic view of the design is shown in **Figure 4**.

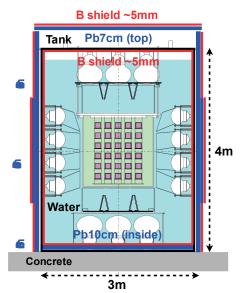


Figure 4. A schematic view of the additional passive shield for the  $(n,\gamma)$  background reduction. The blue and red parts show Pb shield and B-containing rubber sheet, respectively.

We installed Pb shields for  $\gamma$ -rays from neutron captures in rocks, and Si rubber sheets including 40 wt% of B<sub>4</sub>C (6.2 wt% of  $^{10}B$ ) inside and outside the detector to reduce thermal neutron captures in stainless steel tank. The typical Pb thickness is about 10 cm to reduce  $\gamma$ -rays with several MeV to  $\sim\!1/100$ . Considering the weight limit on the top of the detector, Pb thickness in the top was suppressed to 7 cm, but Pb thickness was increased to 12 cm in the center of the detector side, where thickness of the passive water shield in the detector is thinnest. In the bottom, Pb shield was set inside the detector, so as to avoid significant modification of the system.

By installing the shield, the  $(n,\gamma)$  background rate was expected to be ~  $0.7(\pm 50\%)$  events/year/96crystals from the MC simulation. The breakdown of  $(n,\gamma)$  background from rocks and tank is  $0.34\pm 0.14$  and  $0.4\pm 0.2$  events/year/96crystals, respectively.

#### 4.2. Shield construction

The shield construction started in March 2015. The status of the construction is shown in **Figure 5**. Firstly, we started to set Pb shields from the outside of the detector. The bottom Pb blocks were set inside of the tank. They were covered by the liquefied B shield to block Pb dissolution to the water.

The B<sub>4</sub>C containing rubber sheets was set after the Pb construction. They were supported by the punched metal outside and the aluminum frame inside. After setting inner sheets, we poured the liquefied B shield in the bottom of the tank. They were solidified in 1 day.

Finally, we closed the detector after installing Pb shield on the top. The shield construction finished in March 2016 and then we started the data taking. We checked B and Pb elution into water periodically, and no elution has been observed yet.

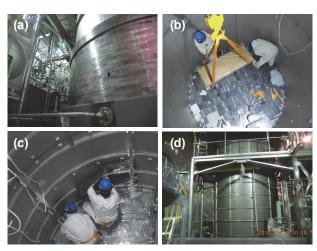


Figure 5. Photos taken in the shield construction. (a) Pb shield outside the tank. (b) Pb shield in the bottom of the tank. (c) B sheet set inside the tank. (d) Overview of the shield.

#### 5. Results

### 5.1 Energy spectrum after shielding

The energy spectrum after  $(n,\gamma)$  shield setup is shown in **Figure 6**. In this case, the LS cut and the <sup>208</sup>Tl cut are not applied to emphasize counting statistics in the high energy region. The red and green histograms show the spectrum before  $(n,\gamma)$  shield setup, and the blue histogram shows the spectrum after setup. All spectra are normalized to the event rate in 56.9 days.

The peaks at 1.46 MeV and at 2.6 MeV caused by environmental  $\gamma$ -rays from  $^{40}$ K and  $^{208}$ Tl, respectively, are reduced by about one order of magnitude by the additional shield. This would help to reduce the dead time when we apply the  $^{208}$ Tl veto. Events in 3 to 4 MeV region were reduced by replacing the O-ring and Al sleeve near the CaF<sub>2</sub> crystals with ones having less  $^{232}$ Th activity.

The events above 5 MeV ( $(n,\gamma)$  background) were clearly reduced by  $(n,\gamma)$  shield setup. The reduction rate is an order of  $10^{-2}$  on average, and consistent with the design value in the MC simulation.

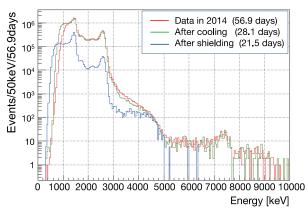


Figure 6. The observed energy spectrum without LS cut and  $^{208}$ Tl cut before and after the  $(n,\gamma)$  shield setup. The red and green histograms show the spectra before setup, and the blue histogram shows the spectrum after setup. All spectra were normalized to the event rate in 56.9 days.

## 5.2 Sensitivity of CANDLES after (n, y) shielding setup

Then, the analysis cuts for  $0\nu\beta\beta$  selection were applied to simulate realistic situation. The analysis was performed for all crystals or pure crystals whose  $^{232}\text{Th}$  activity were below 10  $\mu\text{Bq/kg}$ . The energy spectrum for 27 pure crystals is shown in **Figure 7**. A summary of the analysis result is shown in **Table 1**.

As a conclusion, no events were observed in the region of interest  $(Q_{\beta\beta\text{-}1\sigma}^{+2\sigma};~4170\text{-}4480~\text{keV})$  during the measurement for 21.5 days. From the number of expected backgrounds and the signal efficiency, the sensitivity of the present CANDLES for  $0\nu\beta\beta$  half-life is calculated to be  $0.9\times10^{23}$  year and  $0.5\times10^{23}$  year for all crystals and 27 crystals, respectively. Comparing with the best half-life limit using  $^{48}\text{Ca}$  so far  $(0.6\times10^{23}~\text{year})$ , it will be possible to obtain the better result in a 1 year measurement.

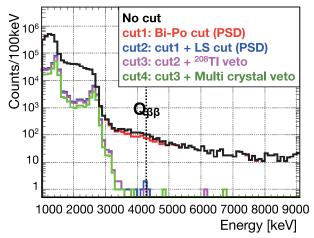


Figure 7. The energy spectrum after  $(n,\gamma)$  shield setup in the 21.5 days live-time measurement by applying various analysis cuts, i.e. the Bi-Po cut, the LS cut, the  $^{208}$ Tl veto, and the multi crystal veto that are described in section 3.2, 2, 3.3, 3.3, respectively. The green histogram shows the spectrum obtained with all analysis cuts. There are no events in the region of interest ( $Q_{\beta\beta-1\sigma}^{+2\sigma}$ ; 4170-4480 keV).

Table 1. Results of with the 0νββ analysis after shield setup.

Crystal selection	All crystals	27 crystals
Number of events	0	0
Expected <sup>208</sup> Tl BG	1.4	0.14
Expected (n, y) BG	0.04	0.01
Signal efficiency	0.30	0.30
Sensitivity in 1 year	$0.9 \times 10^{23} \text{ year}$	$0.5 \times 10^{23} \text{ year}$

#### 6. Conclusion

CANDLES is designed for the study of double beta decay with  $^{48}\text{Ca}$ . The main background sources are  $^{208}\text{Tl}$  decay in  $\text{CaF}_2$  crystal and high energy  $\gamma\text{-rays}$  from neutron captures in surrounding rocks and stainless steel. The latter  $(n,\gamma)$  events were found to be the largest background, and we installed an additional passive shield consisting of Pb and B<sub>4</sub>C rubber sheets. We confirmed that the reduction rate of the  $(n,\gamma)$  background was about  $10^{-2}$ . No events were observed in the region of interest in 21.5 days measurement. The expected sensitivity in 1 year is  $0.9\times10^{23}$  year, which is the best result among the  $0\nu\beta\beta$  measurements with  $^{48}\text{Ca}$ .

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