研究論文

2ν-ZICOS experiment for an observation of two neutrino emission double beta decay using ^{96}Zr nuclei

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Abstract

2ν-ZICOS experiment has been planned to measure the half-life of two neutrino emission double beta decay for ^{96}Zr nuclei. A newly designed 2ν-ZICOS detector will observe about 100 events of ^{96}Zr double beta decay. The detector will use an ultra-pure quartz flask and 20 Hamamatsu H3378-50 photomultipliers in order to distinguish the signal shape of Cherenkov lights. The liquid scintillator containing 0.4 g of ^{96}Zr will be filled in the ETFE cubic bag. The transparency of our ETFE sheet for scintillation light was 97.5% at minimum. The constructions of a radiation shield using Pb blocks and 2ν-ZICOS detector will start in the early fiscal year 2024 at LAB-A in Kamioka mine, and the observation will start in next summer or autumn at the latest.

Key words : Neutrino emission double beta decay, Liquid scintillator,
Zirconium-96, Background reduction, U/Th contamination

1. Two neutrino emission double beta decay using ^{96}Zr

According to some recent results for neutrino physics, a neutrino should have a mass. Direct measurement of the absolute neutrino mass was tried by the KATRIN experiment and the recent result was obtained by $m_\nu < 0.7$ eV/c^2 [1]. Although the cosmological observations will give us the sum of neutrino mass (Σm_ν) as a range of 15 - 50 meV for some combination of next-generation Cosmic Microwave Background and Large Scale Structure observation [2], the neutrinoless double beta decay ($0\nu\beta\beta$) has an ultimate sensitivity to not only the absolute neutrino mass but also the nature of particle physics whether Dirac or Majorana.

The half-life was expressed by following formula;

$$[T^{0\nu}_{1/2}]^{-1} = G^{0\nu}|M^{0\nu}|^2\langle m_{\beta\beta}\rangle^2 \quad (1)$$

where $G^{0\nu}$ is the precisely calculated phase space factor, $M^{0\nu}$ is the nuclear matrix element, and $\langle m_{\beta\beta}\rangle$ is the effective Majorana mass of electron neutrino. Moreover, $0\nu\beta\beta$ violates the lepton number so it should be used for the probe beyond the Standard Model.

In the neutrino oscillation scenario, there are three eigenstates of the neutrino mass $m_{1,2,3}$ and the mixing angle $\theta_{1,2,3}$. The difference of mass eigenstatesquare was determined by the measurements of atmospheric neutrinos (Δm^2_{23}) [3][4][5][6], solar neutrinos(Δm^2_{12}) [7][8], and reactor neutrinos(Δm^2_{13}) [9][10][11], however, the ordering was not fixed yet. Although a normal

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mass ordering (1-2-3) indicates that the probability of discovery for the neutrino mass becomes ultimately zero, an inverted mass ordering (3-1-2) shows the discovery potential should appear around $T_{1/2} \leq 10^{28}$ years or $m_{\beta\beta} \geq 0.02$ eV/c^2.

The half-life is experimentally written by following formula;

$$T^{0\nu}_{1/2} \propto \epsilon\sqrt{\frac{Mt}{B\Delta E}} \tag{2}$$

where ϵ is the detection effciency, M is the mass of target isotope, t is the observation time, B is the background event rate, and ΔE is the detector energy resolution. In order to achieve the half-life to be 10^{28} years, the detector should have a ton scale of target mass at least.

On the other hand, the measurement of two neutrino emission double beta decay ($2\nu\beta\beta$) is quite important for the determination of the matrix element $M^{2\nu}$ which is used for the calculation of $M^{0\nu}$ in some models.

Zirconium-96 (^{96}Zr) is a candidate nuclei of $2\nu\beta\beta$ and Q-value is 3.35 MeV. The half-life was measured by NEMO-3 experiment using the tracking drift chamber. Using 9.4 g of ^{96}Zr and 1221 days observation corresponding to 0.031 kgy, the obtained half-life was $T^{2\nu}_{1/2}$ = [2.35 ± 0.14(*stat.*) ±0.16(*sys.*)] × 10^{19} years [12]. They observed 429.2 ± 26.2 events with a 7.5% effciency. Using this result, about 200 events for $2\nu\beta\beta$ using 1 g of ^{96}Zr will be observed, if the detector effciency is 100%.

2. 2ν-ZICOS experiment

ZICOS is one of the future experiments for $0\nu\beta\beta$. The target nuclei is ^{96}Zr and the Q-value is 3.35 MeV, therefore the radioactive background events such as ^{214}Bi in Uranium series and ^{10}C, which is spallation product of energetic cosmic muons, could be removed by their lower energy.

The conceptional design of ZICOS detector is shown in the left panel of Fig. 1. The detector consists of spherical frame mounted by photomultipliers (PMT), inner balloon filled with 113 tonnes of liquid scintillator containing tetrakis(isopropyl acetoacetato) zirconium (Zr(iPrac)$_4$), and pure Anisole for outside of the inner balloon. Therefore it is almost similar structure as KamLAND-Zen detector. As reported by KamLAND-Zen [13], non-negligible background events were found around 3 - 4 MeV, and those were the decay products from ^{208}Tl which was adhere on the surface of inner balloon. Fortunately, the Q-value of ^{136}Xe is 2.479 MeV so those background events did not affect due to out of region of interest (ROI).

However, those background events should be serious for ZICOS experiment, because of the overlap with ROI

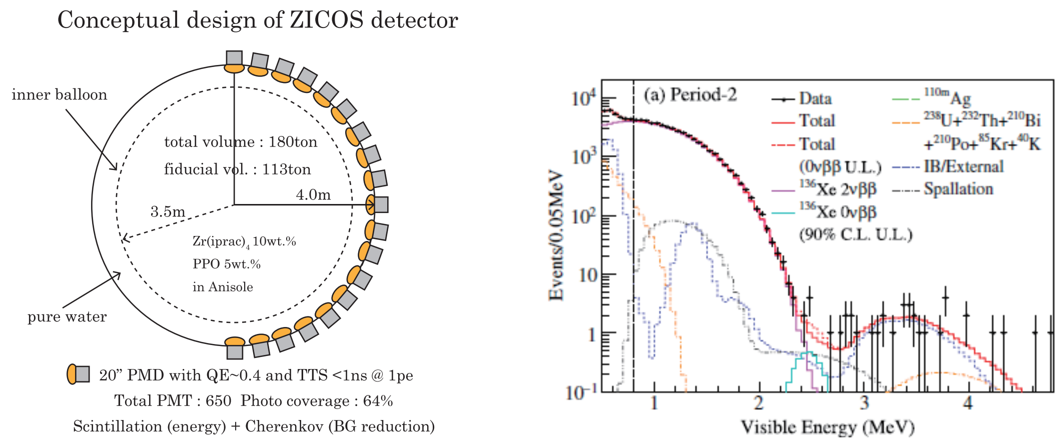

Figure 1. The left side panel shows the conceptional design of ZICOS detector. The inner detector is located in a pure water tank, and has photomultipliers with 64% photo coverage. The inner balloon will be filled with a liquid scintillator which contains 10 wt.% of Zr(iPrac)$_4$, 5 wt.% of PPO and 0.1 wt.% of POPOP. The outside of inner balloon will be filled with a pure Anisole in order to reduce backgrounds. The right side panel shows the results from KamLAND-Zen ^{136}Xe observation [13].

for ^{96}Zr. In order to obtain half-life of order 10^{27} to 10^{28} years, we have to enrich ^{96}Zr to be 20 - 50% and reduce 95% of ^{208}Tl background events as observed in KamLAND-Zen. Those background events could be removed by the averaged angle which is topological information of Cherenkov lights as discussed in our previous papers [14][15][16][17][18], even though ^{208}Tl decay will occur on the surface of inner balloon.

As described in the previous section, 200 events of $2\nu\beta\beta$ will be observed using an order of 1 g for ^{96}Zr nuclei. For the purpose, we are designing 2ν-ZICOS detector as shown in the left side panel of Fig. 2. This detector uses 16cm diameter round bottom flask using an ultra-pure quartz (GE214), and 20 low-background fast rise-time 2inch PMT Hamamatsu H3378-50, which will be used for pulse shape discrimination of Cherenkov lights, are mounted by the designed jig on the flask. 1 little of liquid scintillator loaded 100 g of $Zr(iPrac)_4$, which contained about 0.4 g of ^{96}Zr, will be filled inside of inner bag. The expected number of signals for 2 neutrino double beta decay is about 100 events per year among about 1 million background events. This detector will be located at LAB-A in Kamioka mine.

The left panel of Fig. 3 shows the underground laboratory in Kamioka mine. LAB-A is just behind of LINAC control room, where locates beside of Super-Kamiokande detector. There are some stuffs and booths in LAB-A as shown in the right panel of Fig. 3. At the end section of those booths, there is small space where Pb blocks are stored now. We will install our clean booth (class 1000) here and the radiation shields using these Pb blocks will be prepared. Inside this shield, we will construct 2ν-ZICOS detector in

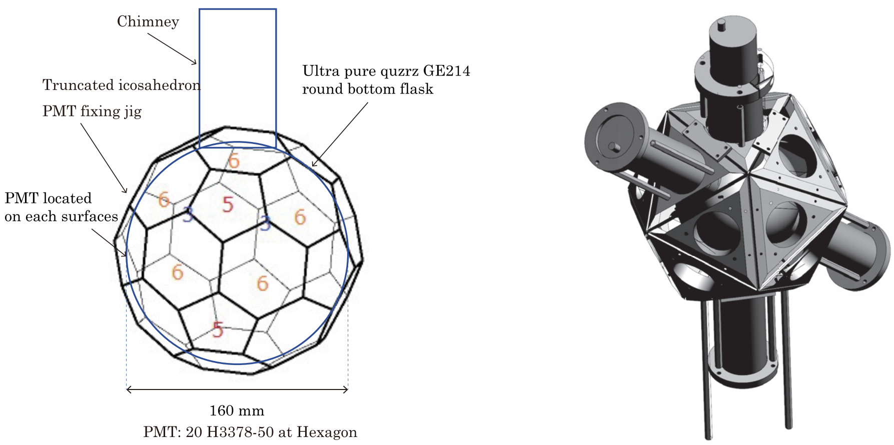

Figure 2. The left side panel shows the designed 2ν-ZICOS detector. The ultra-pure quartz round bottom flask and 20 Hamamatsu H3378-50 photomultipliers will be used for the observation of scintillation lights. The right side panel shows a sample of the PMT mounting jig.

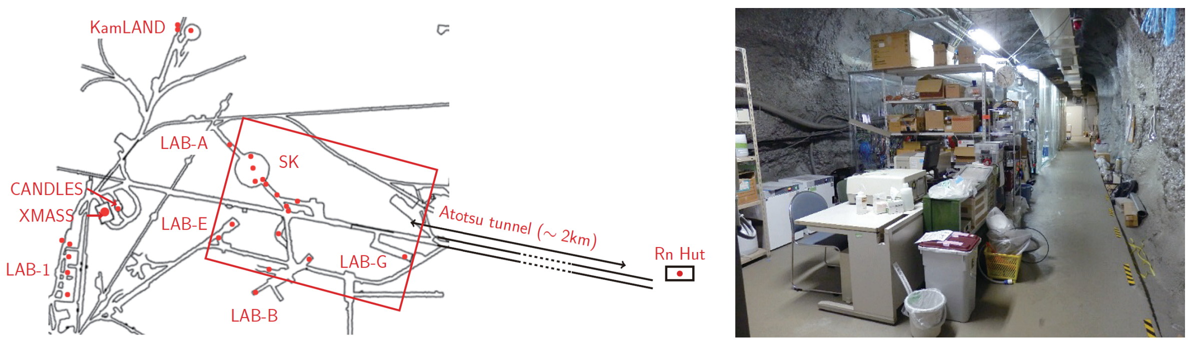

Figure 3. The left side panel shows the map of underground laboratory in Kamioka mine. The right panel shows the photograph of LAB-A.

the early fiscal year 2024.

3. U/Th contamination and simulation

The contamination of Uranium and Thorium for the ultra-pure quartz, $Zr(iPrac)_4$, and Zirconium tetrachloride ($ZrCl_4$) as a material of zirconium complex were measured by the ICP mass spectrometer. We found that most serious backgrounds come from quartz itself. The amounts are 15 ppb for Thorium and 29 ppb for Uranium, respectively. Those amounts correspond to 6×10^{-5} Bq/g for Thorium, and 4×10^{-4} Bq/g for Uranium. Assuming the radiation perpetual equilibrium, the expected number of backgrounds from the quartz are 1 million events for ^{208}Tl and 6 million events for ^{214}Bi per year, respectively.

Assuming those amounts, the energy distribution of each background events and expected signal from double beta decay are simulated by Monte Carlo as shown in Fig. 4. Top, middle, and bottom panels of Fig. 4 show the case of ^{208}Tl, ^{214}Bi, and ^{40}K, respectively. The solid, dashed, and dotted lines show all events, events after fiducial volume cut which means the vertex position within 5 cm from center of the detector, and events after averaged angle cut which means the angle greater than 60 degree, respectively. The hatched spectrum indicated $2\nu\beta\beta$ signal. According to these figures, ^{40}K affects only a part of double beta decay signal. ^{214}Bi is significant background, but a small fraction of double beta decay signal should be observed after the fiducial volume cut and averaged angle cut. However, ^{208}Tl is the most serious background even after those cuts. Only a few events for $2\nu\beta\beta$ might be

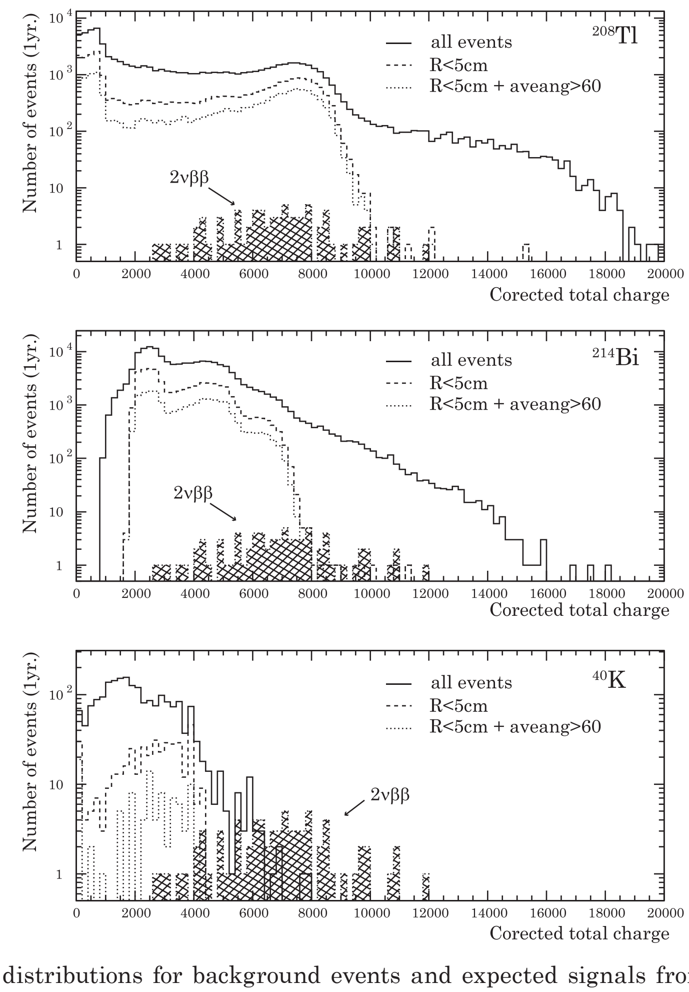

Figure 4. The energy distributions for background events and expected signals from double beta decay are simulated by Monte Carlo. Top, middle, and bottom panels show the case of ^{208}Tl, ^{214}Bi, and ^{40}K, respectively. The solid, dashed, and dotted lines show all events, events after fiducial volume cut, and events after averaged angle cut, respectively. The hatched spectrum indicated ^{96}Zr double beta decay signal.

observed. In this simulation, we used cubic bag for storing scintillator as described in next section.

4. Current status

In this section, a current status of the observation for $2\nu\beta\beta$ is explained. The clean booth (class 1000) was constructed in the Laboratory of Miyagi University of Education as shown in the left panel of Fig. 5 for a preparation of the liquid scintillator and a mock up of 2ν-ZICOS detector. 200 g of $Zr(iPrac)_4$ has been synthesized by NARD Institute, Ltd., and it is stored in the dry box located inside of the clean booth. Inside of the clean booth, the glove box with N_2 gas circulation for the preparation of liquid scintillator has been also installed as shown in the left panel of Fig. 5. 12 Hamamatsu H3378-50 PMTs have been already arrived in last year, and stored in the clean booth. Remaining 8 PMTs were just delivered in September 2023.

The special made round bottom flask using the ultra-pure quartz has been produced by Asahi Glassplant Inc., and it is also stored in the clean booth. Both PMT mounting jig and the inner bag for storing liquid scintillator are being designed now. The middle panel of Fig. 5 shows the special made round bottom flask. The chimney could be used for PMT guide to detect scintillation.

We will use the ETFE transparent sheet (50 μm) for inner cubic bag to store liquid scintillator as shown in the right panel of Fig. 5. Outside of inner bag, pure Anisole will be filled in the detector. The ETFE is a kind of Fluoro resin against Anisole erosion, and no damage due to Anisole erosion was found in one month test. However, the welding of Fluoro resin is typically so diffcult that the company, which has some productions of the PTFE bag, is now testing the welding using the ETFE sheet.

The transparency of the ETFE sheet was directly measured by the scintillation lights from liquid scintillator. The left side panel of Fig. 6 shows a setup for the transparency measurement. We used a special light guide between the 120 mL vial containing liquid scintillator and 4 PMTs (Hamamatsu H6410) as shown in the right panel of Fig. 6. 3 PMTs were mounted for side of the vial, and 1 PMT was for bottom. The radio-active source ^{137}Cs was located on top of the vial, and gammas irradiated the liquid scintillator. The trigger signal was made by coincidence of signals from all PMTs, and the data was taken by the charge sensitive ADC (LeCroy 1182).

Using photon peak with or without the ETFE sheet covered side of the vial which is shown in the right panel of Fig. 6, the transparency using the side PMT light yield could be calculated the ratio of peak position. For the consistency check, the bottom PMT

Figure 5. The left panel shows a clean booth and the grove box. The middle panel shows the specially made round bottom flask using the ultra-pure quartz. The chimney could be used for PMT guide. The right panel shows the figure of the inner cubic bag which has 10 cm in one side

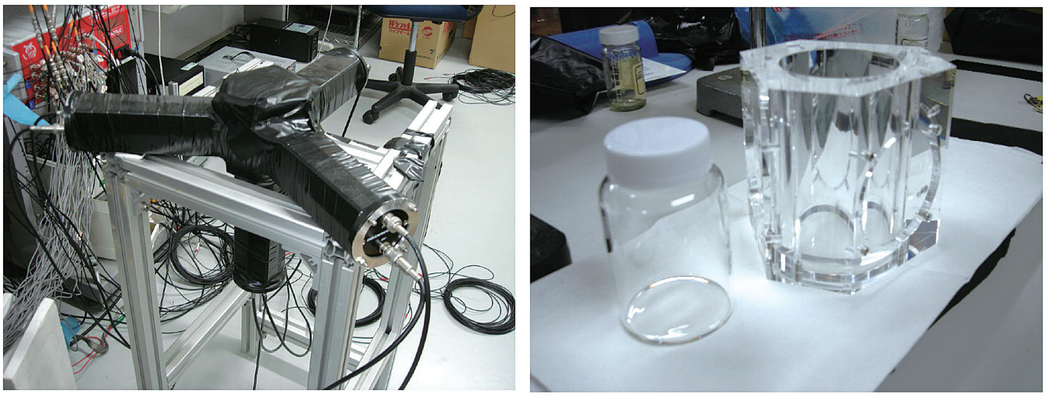

Figure 6. The left panel shows the setup for the measurement of a transparency. The ETFE sheet was covered by side of the 120 mL vial as shown in the right panel.

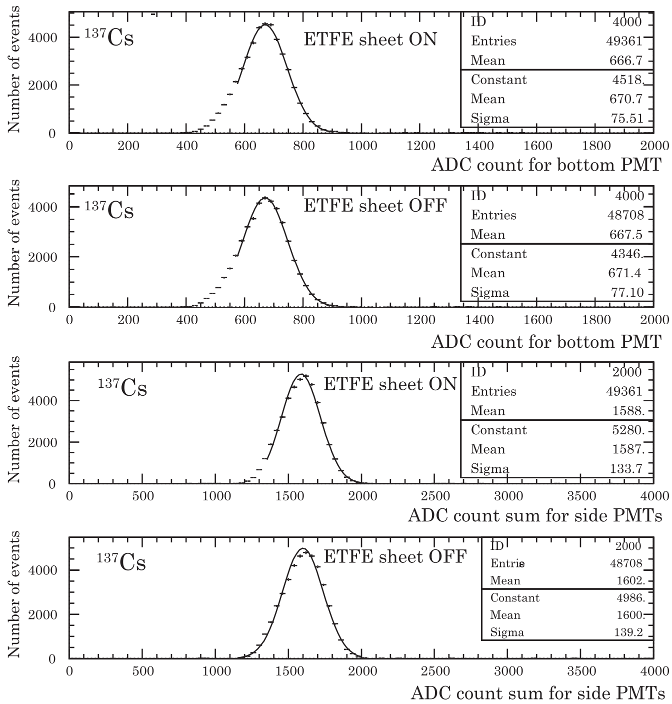

Figure 7. The charge distribution of photo peak for bottom PMT and side PMTs with of without the ETFE sheet using ^{137}Cs 662 keV gamma.

should measure same photon yield, because of no ETFE sheet inserted for both cases.

Figure 7 shows the energy spectra measured by ^{137}Cs. The top two figures show in case of bottom PMT, and the bottom two figures shows in case of side PMTs. The ratio of peak position is 1.0015 ± 0.0018 for bottom PMT and it is consistent with 1.0 within the statistical error. This means that the same light yield was observed as expected. On the other hands, the ratio of peak position for the case with and without the ETFE sheet for side three PMTs was obtained by 0.9768 ± 0.0014, and only 2.5% light yield loss should be expected at maximum due to the ETFE sheet.

5. Enrichment of ^{96}Zr

Finally, we would like to discuss the Centrifuge for ^{96}Zr isotope separation. The Centrifuge plant of Japan Nuclear Fuel Ltd is located at Rokkasho village in Japan, and there are a lot of Centrifuge towers in their plant. (See their homepage)

We have discussed with the staff members of Technical

Development Center for Uranium Enrichment for the isotope separation of ^{96}Zr using their Centrifuge plant. We proposed that $ZrCl_4$ is one of the choices for the gas, because of low sublimation point 332 degree Celsius at atmospheric pressure. Of course, $ZrCl_4$ is a material of $Zr(iPrac)_4$, therefore no additional chemical process needs for the synthesis of $Zr(iPrac)_4$.

Using a natural abundance combination for $ZrCl_4$, there are 13 molecular masses, since 5 isotopes for Zirconium and 2 isotopes for Chlorine naturally exist. Figure 8 shows the number of molecules existence for each combination in case of total 100,000 molecules. Top two figures show the all molecules and molecules including ^{96}Zr. In this case, a ratio of the selected molecules corresponds to 2.8% of ^{96}Zr, in other words, the natural abundance of ^{96}Zr was reproduced. On the other hand, if the isotope separation was made by the Centrifuge as molecular mass 238 or more, then a ratio of the separated molecules included ^{96}Zr is 26.2% as shown in bottom two panels of Fig. 8. Therefore 26% enrichment maybe possible, if the Centrifuge can be used for $ZrCl_4$.

6. Future plan

The preparation of 2 little of ZICOS liquid scintillator will start in October 2023. The radiation shield using Pb blocks with the falling prevent wall is now designed. The inner size may be 80 cm cubic. Tuned inner bag using the ETFE sheet will be ready after welding test by the company. Mock up for PMT mounting jig with flask will be done in the early 2024. After that, the installation of inner bag and bringing the liquid scintillator into the bag will be tested by using a ultra-pure water, not a scintillator in order to avoid the risk. The construction of the Pb shield and 2ν-ZICOS detector will start in the early fiscal year 2024 in LAB-A, and the observation will start in next summer or autumn at the latest.

Above plan will be done by 3rd year undergraduate students of Miyagi University of Education as a part of the graduation research.

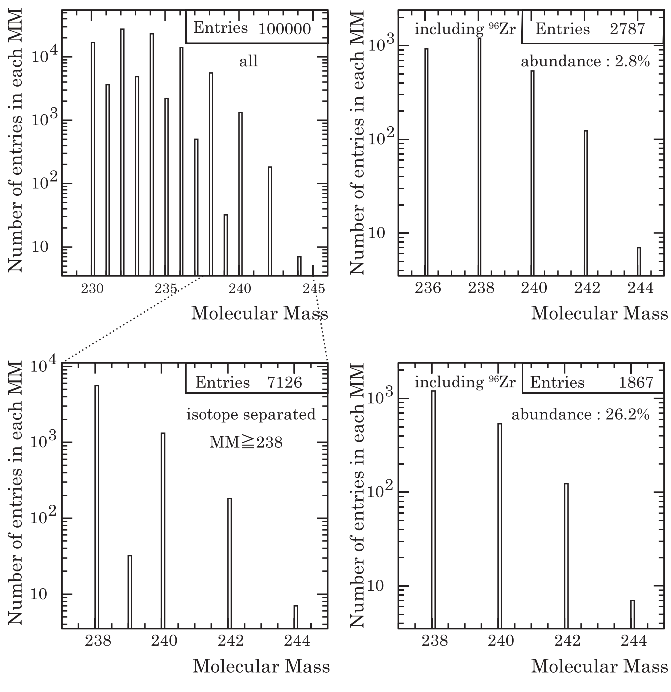

Figure 8. $ZrCl_4$ has 13 molecular masses using natural abundance combination. The top two panels show number of molecules for all case (100,000) and molecules including ^{96}Zr, respectively. Comparing those number, 2.8% was obtained and it is consistent with natural abundance of ^{96}Zr. The bottom two panels show that the number of molecules which is separated by Centrifuge with molecular mass 238 or more, and molecules included ^{96}Zr. In this case, 26.2% enrichment might be possible.

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（令和６年１月30日受理）

^{96}Zr原子核を用いたニュートリノの放出を伴う二重ベータ崩壊の観測のための2ν-ZICOS実験

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要　旨

2ν-ZICOS実験は、^{96}Zr 原子核を用いて２つのニュートリノを放出する二重ベータ崩壊の半減期を観測する実験である。新たにデザインされた２ν-ZICOS 検出器は、約100事象の二重ベータ崩壊を観測する計画である。この検出器は、高純度石英ガラスによる特製丸底フラスコに、波形を用いてチェレンコフ光の有無を判別することができる20本の浜松ホトニクス製H3378-50光電子増倍管を設置している。検出器内には、0.4g の ^{96}Zr を含む1リットルの液体シンチレータをETFE 製立方体バックの中に格納している。ETFE のシンチレーション光の透過率は、97.5% 以上である。鉛ブロックによる遮蔽体と２ν-ZICOS 検出器の建設は、2024 年度初頭に神岡地下観測所内のLAB-A 内で開始し、夏または遅くとも秋には観測を開始する計画である。

Key words：ニュートリノの放出を伴う二重ベータ崩壊，液体シンチレータ，
ジルコニウム96，背景事象除去，ウラン・トリウムによる汚染

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