ZICOS : New project for neutrinoless double beta decay experiment using zirconium complex in liquid scintillator

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Abstract

A liquid scintillator containing a zirconium β-keto ester complex has been developed for the Zirconium COmplex in liquid Scintillator (ZICOS) experiment which is new project of neutrinoless double beta decay search. We are aiming to develop a detector which has a good energy resolution (3.5% at 3.35 MeV), a large light yield (60% that of BC505) and a low background rate (0.1 counts/tonne・year) with several hundred kg of ^{96}Zr isotope, so we have investigated the zirconium β-keto ester complexes such as tetrakis (isopropyl acetoacetato) zirconium and tetrakis (ethyl acetoacetato) zirconium, which have high solubility (over 30 wt.%)in anisole. We measured the performance of liquid scintillator containing these zirconium β-keto ester complexes, and obtained 48.7±7.1% of the light yield of BC505 and the energy resolution of 4.1±0.6% at 3.35 MeV assuming 40% photo coverage of the photomultiplier in the detector. This results reached our initial goal, so we estimated that ZICOS experiment should be sensitive to $\langle m_\nu \rangle < 0.2 - 0.3$ eV assuming g_A = 1.25, g_{pp} = 1.11 and QRPA model, if a radius of the balloon is 1.5 m and the balloon is filled with a liquid scintillator containing 10 wt.% concentration of a zirconium β-keto ester complex with an enriched ^{96}Zr.

Key words：Neutrino （ニュートリノ）

Neutrinoless Double Beta Decay （ニュートリノを放出しない２重ベータ崩壊）

Majorana Mass (マヨラナ質量)

Zirconium （ジルコニウム）

Liquid Scintillator （液体シンチレータ）

1. Introduction

Recent results from long baseline neutrino experiments [1] and reactor neutrino experiments [2] have confirmed a non-zero θ_{13}, and reliable neutrino mass matrix elements, except for the mass hierarchy and CP phase in the lepton sector, can now be obtained by combining results from all observations of atmospheric [3], solar [4], and reactor neutrinos [5]. However, neither the Standard Model nor SUSY extensions of it explain why neutrinos should have a mass.

On the other hand, recent leptogenesis models postulate the existence of heavy right-handed neutrinos with masses as large as the Planck scale [6], which also generally present in the See-Saw model [7], and strongly favor the existence of Majorana neutrinos. The observation of

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neutrinoless double beta decay would confirm the Majorana nature of the neutrino and would also provide more information about the neutrino mass scale and the mass hierarchy.

Neutrinoless double beta decay $(0\nu\beta\beta)$ is expected to occur in special nuclei, such as those listed in Table 1. These nuclei cannot undergo single beta decay into the nucleus with an atomic number one higher because of its larger mass. However, if the nucleus with an atomic number two higher has a lower mass than the original nucleus, then double beta decay could occur. In ordinary double beta decay $(2\nu\beta\beta)$, two electrons and two neutrinos are emitted at the same time. In this case, the total energy of the two electrons should be spread out between 0 and the Q-value, because the two neutrinos would take out some energy. On the other hand, $0\nu\beta\beta$ does not emit neutrinos (emits only two electrons), because the neutrino emitted from one nucleon should be absorbed by another nucleon in same nucleus due to the Majorana nature of the neutrino. Therefore, the total energy of the two electrons should be unique and be the same as the Q-value of the initial and final nuclei.

To determine the Majorana neutrino mass from $0\nu\beta\beta$, we must measure the half-life. The half-life of $0\nu\beta\beta$ is given by

$$[T_{1/2}^{0\nu}(0^+ \to 0^+)]^{-1} = G_{0\nu}M_{0\nu}^2 \frac{\langle m_\nu \rangle^2}{m_e^2} \qquad (1)$$

Isotope (decay process)	Q-value (keV)	Abundance (%)
^{48}Ca $\to ^{48}$Ti	4271	0.2
^{150}Nd $\to ^{150}$Sm	3667	6
^{96}Zr $\to ^{96}$Mo	3350	3
^{100}Mo $\to ^{100}$Ru	3034	10
^{82}Se $\to ^{82}$Kr	2995	9
^{116}Cd $\to ^{116}$Sn	2802	7
^{130}Te $\to ^{130}$Xe	2533	34
^{136}Xe $\to ^{136}$Ba	2479	9
^{124}Sn $\to ^{124}$Te	2288	6
^{76}Ge $\to ^{76}$Se	2040	8
^{110}Pd $\to ^{110}$Cd	2013	12

Table 1. Possible nuclei isotopes for $0\nu\beta\beta$.

where $G_{0\nu}$ is the kinematic phase space factor, $M_{0\nu}$ is the matrix element of the target nuclei including Fermi, Gamow-Teller and tensor contributions, m_e is the electron mass, and $\langle m_\nu \rangle$ is the effective Majorana neutrino mass. According to Eq.(1), we must be able to measure a half-life of the order of 10^{25} years assuming the neutrino mass to be below 0.1 eV. On the other hand, the half-life can also be expressed experimentally as

$$[T_{1/2}^{0\nu}]^2 \sim a\frac{MT}{\Delta E\, B} \qquad (2)$$

where a is the abundance of the target isotope, M is the target mass, T is the measurement time, ΔE is the energy resolution, and B is the background rate. According to Eq.(2), next-generation $0\nu\beta\beta$ experiments should have tonnes of target isotope, a background rate of 0.1－1 counts/(tonne・year), and an energy resolution of 3.5% at 3.35 MeV. (Alternatively we could combine a relatively low target mass with very high energy resolution.)

2. ZICOS experiment

We are going to search for $0\nu\beta\beta$ signal using nucleus ^{96}Zr in the liquid scintillator. This experiment is named Zirconium COmplex in liquid Scintillator (ZICOS) torneutrinoless double beta decay experiment. The liquid scintillator will be contained in inner detector. If the radius of inner detector is 1.5 m, then total volume is 14.1 m^3. This detector is located in a cylindrical tank as shown in Fig 1, which is 5.4 m in diameter and is 5 m in height, assuming EGADS tank[8]. This tank should be filled with a pure mater in order to exclude external γ rays and neutrons. Photomultiplier will be mounted on the wall of both the inner detector and the cylindrical tank. The photo coverage of inner detector should be 40% in order to collect scintillation light efficiently. To eliminate cosmic muons, the detector should be located in an underground laboratory, such as the Kamioka Observatory.
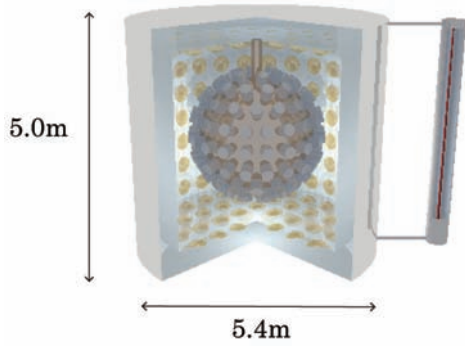
Figure 1. The conceptual design of the ZICOS detector. The liquid scintillator is contained inside a balloon, which is surrounded by a pure mater to reduce external backgrounds.

A nucleus of ^{96}Zr has a Q-value of 3.35 MeV, which is the third largest value in possible double beta decay nuclei, and 3 % natural abundance, as shown in Table.1. Generally speaking, it is difficult to use metals, such as ^{100}Mo[9], ^{150}Nd[10], ^{96}Zr[11] and ^{82}Se[12] in a double beta decay experiment. Some metal nuclei such as ^{76}Ge[13], ^{116}Cd[14], and ^{130}Te[15] could be used for solid-state detectors or scintillation crystals. These detectors should have very good energy resolution, however, the target mass is limited by the size of the detector. Usually the metals are used as a thin foil target and the emitted electrons are tagged by the magnetic field in the track chamber like NEMO-3 experiment[9][10][11][12]. The curvature of the trajectory corresponds to the electron energy. In that case, the energy loss in the foil might not be negligible and anyway in such detectors it is generally difficult to have a tonne of target. On the other hands, a nucleus ^{136}Xe has gaseous and liquid phase, so that suitable detector could be considered. Recently, the high pressure of Xe gas has been proposed to use not only for the tracking chamber but also for the scintillation detector [16]. Although a liquid xenon is high light yield scintillator, it could be used for the time projection chamber not only as a target but also as a detector. A recent result shows that the half-life of ^{136}Xe is obtained as $T_{1/2} > 1.6 \times 10^{25}$

years at 90 % confidence level [17].

On the one hand, KamLAND-Zen have been succeeded to dissolve enriched ^{136}Xe gas into a liquid scintillator with 320kg, and they obtained limits of life time as $T_{1/2}^{0\nu} > 1.1 \times 10^{25}$ years [18]. This indicated that a liquid scintillator should have the scalability to realize tonnes of the target, and will be easy shield from external backgrounds by being located in a tank. Although novel techniques, such as using quantum dots to dissolve metals in the liquid scintillator, have been developed in recent years, the solubility is only of the order of 0.1 wt.% [19]. We have, therefore, chosen to dissolve a metal in the liquid scintillator using a β-diketone complex. A classical liquid scintillator consists of an organic solvent such as benzene, toluene or 1,2,4-trimethylbenzene. We choose an anisole (methoxybenzene) as a solvent, because it can dissolve a metal complex of acetylacetone at over 10 wt.%. Our standard scintillator cocktail is produced by dissolving 100 mg PPO (2,5-Diphenyloxazole) and 10 mg POPOP (1,4-bis(5-phenyloxazol-2-yl) benzene) in 20 mL anisole. This standard cocktail has almost same light yield as BC505, as shown in Fig. 2, and a fast decay time ($\sim$ 20 ns).
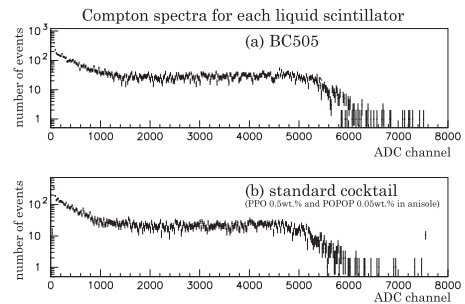

Figure 2. The Compton spectra from a ^{60}Co source for (a) BC505 and (b) the standard cocktail (100 mg PPO and 10 mg POPOP in 20 mL anisole).

The attenuation length of scintillation light ($\sim$ 420 nm emitted from POPOP) measured for anisole is about 6 m as shown in Fig. 3. Although this value is not enough to transmit the scintillation light to photomultiplier, the attenuation length

will be modied around 10 to 15 m by purifying anisole using activated Al_2O_3 as described in Ref. [20].

As shown in Table.1. there are several nuclei which will possibly undergo neutrinoless double beta decay. A β-diketone ligand could be formed as complex with one of these metal nuclei at the coordinate bond. Therefore, the number of ligand bonding with metal nuclei is basically determined by the number of valence electrons which are not used for the covalent bond. For instance, zirconium has three valence electrons. cadmium has two and neodymium has three. Molybdenum has some compounds with 2, 3, 4, 5, or 6 valence electrons. Because of the structure of ligand and the molecular size, it is not easy to understand which is the best metal for formation of the complex. However, we have found that zirconium acetylacetone has the same solubility (over 10 wt.%) as indium acetylacetone, as reported in Ref. [21] Unfortunately, this β-diketone complex has an absorption peak around 290 nm, which deeply overlaps the emission spectrum of anisole, as shown in left panel of Fig. 4. Therefore, it causes color quenching as the amount of dissolved complex is increased [22]. To avoid this quenching, we synthesized a β-keto ester complex instead of a β-diketone complex in order to shorten the absorption wavelength.

According to the absorption spectra for β-keto ester ligand, the peaks were found around 240nm [22].

3. Zirconium β-keto ester complex and its property

Zirconium β-keto ester complex such as a tetrakis (isopropyl acetoacetato) zirconium ($Zr(iprac)_4$) and a tetrakis (ethyl acetoacetato) zirconium ($Zr(etac)_4$) are not commercial products, therefore we have to synthesize them by ourselves.

Tetrakis (isopropyl acetoacetato) zirconium was synthesized by following procedure. $ZrCl_4$
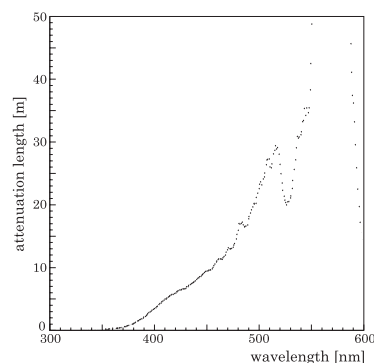

Figure 3. Measured attenuation length in anisole as a function of wavelength.

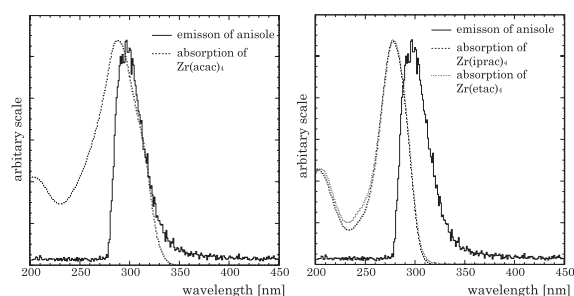

Figure 4. Left panel shows the absorbance spectrum of zirconium acetylacetone (dotted line) and the emission spectrum for anisole (solid line) measured in acetonitrile. Right panel shows the absorbance spectra for tetrakis (isopropyl acetoacetato) zirconium (dotted line) and tetrakis (ethyl acetoacetato) zirconium (dashed line) and the emission spectrum for anisole (solid line).

(3.10 g, 13.3 mmol) in benzene (40 mL) was added to tetrahydrofuran (2 mL), and stirred overnight. The slight cloudy solution was added to isopropyl acetoacetate (8.63 g, 59.9 mmol), and stirred for 1 h at ambient temperature. The pale yellow suspension was added dropwise to triethylamine (5.96 g, 58.9 mmol) over 30 min and then refluxed for 2 h. After cooling, the mixture was filtered to remove amine salt and volatile compounds were removed using a rotary evaporator. The orange residue was recrystallized from isopropyl alcohol. The crystal was collected by filtration and dried under reduced pressure at room temperature to provide $Zr(iprac)_4$ as white crystals (4.63 g, 52.4%). Tetrakis (ethyl acetoacetato) zirconium was also synthesized by following procedure. $ZrCl_4$ (3.94 g, 16.9 mmol) in benzene (50 mL) was added to tetrahydrofuran

(4 mL), and stirred overnight. The slight cloudy solution was added to ethyl acetoacetate (8.86 g, 68.1 mmol), and stirred for 1 h at ambient temperature. The pale yellow suspension was added dropwise to triethylamine (7.03 g, 69.4 mmol) over 30 min and then refluxed for 2 h. After cooling, the mixture was filtered to remove amine salt and volatile compounds were removed using a vacuum pump. A yellow viscous liquid was obtained, which gave pale yellow solid after sitting for several weeks at room temperature. The solid was collected by filtration, dissolved in a small amount of EtOH, and then cooled to -30℃. After 1 month, $Zr(etac)_4$ was obtained as a white solid (8.10 g, 79%).

A chemical structure formula of those complex are shown in Fig. 5, and the chemical formula of those complex are $Zr(CH_3CCOCHCOOCH(CH_3)_2)_4$ (MW=663.87) and $Zr(CH_3CCOCHCOOCH_2CH_3)_4$ (MW=607.76), respectively. Those complexes were white powder or lump as shown in left panel of Fig.6. We measured the solubility of these complexes in anisole and they were over 31.2 wt.% and 32.7 wt.%, respectively. This corresponds to 70g/L for the solubility of Zr metal. This quite high values makes us to realize tons scale detector with small size of detector for low backgrounds and systematic. Indeed a quit transparent scintillator cocktail even for the 10 wt.% concentration of $Zr(iprac)_4$ was obtained as shown in right panel of Fig.6. We also measured the absorbance spectra and they are shown in the right panel of Fig. 4.
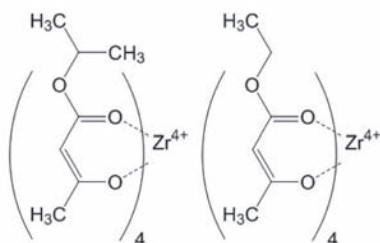

Figure 5. A chemical structure formula of a tetrakis (isopropyl acetoacetato) zirconium (left panel) and a tetrakis (ethyl acetoacetato) zirconium (right panel).

Both absorption peaks of $Zr(iprac)_4$ and $Zr(etac)_4$ were found at 278nm, which shifted only 10 nm to shorter wavelength than that of $Zr(acac)_4$. This was too small to modify the performance of liquid scintillator as we expected by ligand. However, the absorbance spectra of $Zr(iprac)_4$ and $Zr(etac)_4$ became much narrower than that of $Zr(acac)_4$. In other words, there existed less overlap between the absorption spectrum of the β-keto ester complex and the emission spectrum of anisole. Therefore, we could expect the better performance of liquid scintillator even if zirconium β-keto ester complex dissolved with higher concentration.

4. Performance of liquid scintillator containing zirconium β-keto ester complex

The performance of a liquid scintillator from the point of view of neutrinoless double beta decay should be evaluated by its energy resolution. To distinguish between $2\nu\beta\beta$ and $0\nu\beta\beta$ and avoid energetic γ rays from [214]Bi and [208]Tl which are the progeny of [238]U and [232]Th chains, our initial goals should be that (a) the light yield should be larger than 60% that of BC505, and (b) the energy resolution should be 3.5% at 3.35 MeV for a 10 wt.% concentration of zirconium β-keto ester complex.

To measure the light yield and the energy resolution, we must use not only the Compton edge but also the single energy peak obtained by using the back scattering method. Fig. 7 shows the conguration of the ^{60}Co radioactive source, the liquid scintillator containing zirconium β-keto ester complex, and a NaI scintillator. To select the scattering angle of 150 degrees, we collimated the γ rays using lead blocks, as shown in the left panel of Fig. 7. The right panel of Fig. 7 shows typical pulse height distributions of the liquid scintillator and the energy spectrum of NaI scintillator. We could see the single peak in both detectors. The calculated energy of scattered γ rays obtained from the scattering angle was 224

Figure 6. Photograph of synthesized complexes (left panel) and a scintillator cocktail consists of Zr(iprac)$_4$ 2222mg, PPO 993mg and POPOP 10mg in 20mL anisole (right panel).

keV, and the fitted value was 221 keV; therefore the experimental measurements should be correct.

We measured samples with several concentrations of zirconium β-keto ester complex. The left panel of Fig. 8 shows the measured light yield fraction for the standard cocktail as a function of the concentration of Zr(iprac)$_4$ and Zr(etac)$_4$. For comparison, the light yield fraction for Zr(acac)$_4$ is also shown in same figure. The concentration of Zr(acac)$_4$ was equivalent to the concentration of Zr(iprac)$_4$, because the light yield depends on an amount of complex molecules as described below. The light yield of Zr(iprac)$_4$ and Zr(etac)$_4$ are almost same for all concentrations, and they are almost twice larger than that of Zr(acac)$_4$ at 2.5 wt.% concentration. This is because of less overlap region between the absorption spectra of zirconium β-keto ester complex and the emission spectrum of anisole. The light yield of Zr(iprac)$_4$ and Zr(etac)$_4$
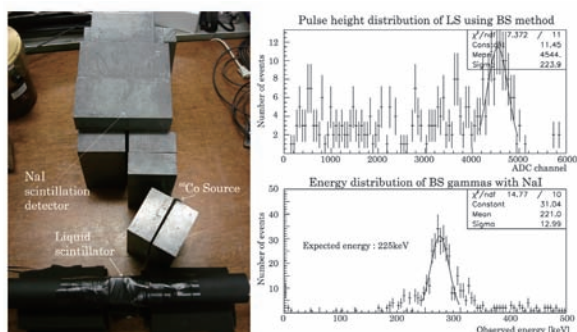

Figure 7. Setup for measurement of the single peak for the liquid scintillator using the back scattering method (left panel) and observed pulse height distribution of liquid scintillator (right top) and the energy spectra of scattered γ rays in the NaI scintillator (right bottom).

could be fitted by using following Eq.(3).

$$\text{Light yield fraction} = \frac{\eta_1 N_{PPO}}{\eta_1 N_{PPO} + \eta_2 N_{Zr}} \qquad (3)$$

Here, η_1 and η_2 show the absorbance of PPO and zirconium β-keto ester complex per mole, and N_{PPO} and N_{Zr} show the amount of PPO and zirconium β-keto ester complex molecules in mole unit. We assume the emission photon could be absorbed by both PPO and zirconium β-keto ester complex with a probability in proportion to the number of molecular and the absorbance, because each absorption spectral shape of PPO and zirconium β-keto complex are almost similar. According to the fitted line in Fig. 8 using Eq(3), the light yield fraction to the standard cocktail for a zirconium β-keto ester complex is expected to be almost 15% at a 10 wt.% concentration. This number is quite smaller than our initial goal.

The right panel of Fig. 8 shows the measured energy resolution as a function of the concentration of Zr(iprac)$_4$ and Zr(etac)$_4$. It appears that the energy resolution obeys the usual expectation $\sigma = \frac{\sigma_0}{\sqrt{E/E_0}}$, where E, E_0, and σ_0 correspond to the electron energy, the reference energy, and the energy resolution for the reference energy, respectively. The energy should be proportional to the light yield so that we used same relation of Eq.(3) as a light yield of each concentration. The obtained energy resolution around 10 wt.% concentration was 35% at 1.03 MeV, which was estimated by measuring the energy of NaI. In this case, the observed energy in the liquid scintillator was 1.03 MeV, because we used ^{60}Co as the γ source (1.33 MeV and 1.17 MeV) and the expected energy deposited in the liquid scintillator would be calculated as 1.03 MeV (the energy of 0.225 MeV observed in the NaI scintillator should be subtracted).

In this setup, the scintillation photons were collected by two photomultiplier, as shown in left panel of Fig. 7. The photo coverage of this setup was estimated to be about 9.3 % using Monte
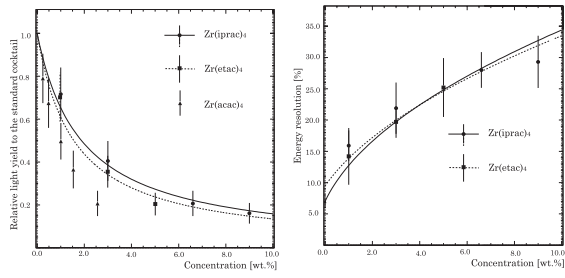
Figure 8. The left panel shows the measured light yield fraction to the standard cocktail as a function of the concentration of $Zr(iprac)_4$ and $Zr(etac)_4$. The right panel shows the measured energy resolution as a function of the concentration of $Zr(iprac)_4$ and $Zr(etac)_4$.
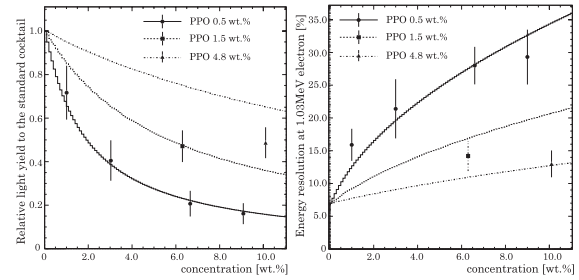

Figure 9. The left panel shows the measured light yield fraction to standard cocktail as a function of the concentration of $Zr(iprac)_4$ in case of the concentration of PPO as 0.5 wt.%, 1.5 wt.% and 4.8 wt.%. The right panel shows the measured energy resolution as a function of the concentration of $Zr(iprac)_4$ in case of same concentration of PPO. Both light yield and energy resolution are recovered by increasing the concentration of PPO with an order of 5 wt.%

Carlo simulation. On the other hand, the ZICOS detector will have 40 % photo coverage of the photomultiplier, so that the energy resolution for the ZICOS detector should be 9.4 % at 3.35 MeV. This value is also quite larger than our initial goal. Therefore, we have to improve the liquid scintillator system in order to get both larger light yield and better energy resolution.

5. Improvement of liquid scintillator system

Observed energy resolution was too large to use for the $0\nu\beta\beta$ decay experiment, even though we ignore the photon attenuation for an actual size of the detector. As we discussed in section 1, we need 3.5% at 3.35 MeV which is Q-value of ^{96}Zr double beta decay at least. This value was estimated by present energy resolution of KamLAND-Zen (4 % at 2.5MeV). In order to reject or identify the backgrounds, especially the signals from $2\nu\beta\beta$ decay, we have to set the energy resolution as low as practical we can.

To improve the energy resolution, the liquid scintillator system should have larger light yield, for instance, 60 % of the BC505. According to Eq.(3), the light yield is in proportion to fraction of the amount PPO molecules with respect to the amount of zirconium β-keto ester complex molecules. In other words, we will be able to modify the light yield of our liquid scintillator system, if we add

more PPO in samples. The dotted and dashed lines in the left panel of Fig. 9 show the expected light yield fraction as a function of the concentration of $Zr(iprac)_4$ using above equation in the case of PPO 1.5 wt.% and 4.8 wt.%, respectively. Also the energy resolution will be modied by same equation for the energy term in the equation $\sigma = \frac{\sigma_0}{\sqrt{E/E_0}}$ as shown in the right panel of Fig. 9. Actually, if we use the concentration of PPO as 1.5 wt.% instead of 0.5 wt.% in 6.3 % concentration of $Zr(iprac)_4$, then the light yield fraction and the energy resolution recovered 47.1 $\pm$ 7.3% and 14.2 $\pm$ 2.3%, respectively. For 10 wt.% concentration of $Zr(iprac)_4$, those values are also modied 48.7 $\pm$ 7.1% and 13.0 $\pm$ 2.0%, respectively, if we use the concentration of PPO as 4.8 wt.% . This energy resolution should correspond to 4.1 $\pm$ 0.6% at 3.35 MeV assuming 40 % photo coverage. Therefore, we could really improve the performance liquid scintillator containing 10 wt.% of zirconium β-keto ester complex with 5 wt.% of PPO, even if the complex made both the light yield and the energy resolution worth due to the photon absorption.

6. Sensitivity of ZICOS experiment for $0\nu\beta\beta$

An experimental result of $0\nu\beta\beta$ for ^{96}Zr was

obtained by NEMO-3 experiment [11]. A lower limit of the life-time was $T_{1/2}^{0\nu} > 9.2 \times 10^{21}$ years and a upper limit of the neutrino effective mass was also $\langle m_\nu \rangle < 7.2 - 10.8$ eV, if nuclear parameters of g_A = 1.25 and g_{pp} = 1.11, and the nuclear matrix model QRPA were used.

As described in section 2, we are planning to use 1.5 m radius spherical inner detector which will be filled with the liquid scintillator containing $Zr(iprac)_4$. The total volume will be 14.1 cubic meters and the weight corresponds to 14.1 tonnes because the density of an anisole is 0.995 g cm^{-3}. Assuming 10 wt.% concentration of $Zr(iprac)_4$, the amount of zirconium should be 216 kg which includes 6.5kg of ^{76}Zr. This corresponds to 9.2 kg of ^{136}Xe which means that 0.03 times to KamLAND-Zen[18]. Assuming same energy resolution, background rates, and the measurement time as those of KamLAND-Zen, we can estimate the sensitivity for the lifetime measurement as $T_{1/2}^{0\nu}$ > 4.4×10^{24} years. This is not enough for $0\nu\beta\beta$ experiment.

In order to increase sensitivity, we have to use another improvements. One method is an enrichment. NEMO-3 experiment used 7g of ^{96}Zr with an enriched to 57.3 % for their target [23]. If we can use 58.5 % enrichment of ^{96}Zr, which is a commercial grade, then the amount of ^{96}Zr will be 126 kg. This corresponds to 0.56 times ^{136}Xe 320 kg of KamLAND-Zen, and the lifetime limits is obtained by $T_{1/2}^{0\nu}$ > 1.1×10^{25} years. This is quite 1155 times longer than NEMO-3 limits. Using Eq.(2), it corresponds to $\langle m_\nu \rangle < 0.2 - 0.3$ eV assuming same parameters of g_A = 1.25, g_{pp} = 1.11 and QRPA model.

To look for the neutrino mass below 0.1eV, we have to decrease backgrounds around 3.5MeV region at one order magnitude less than KamLAND-Zen. According to recent analysis of KamLAND-Zen, they found that those backgrounds consist of decay products from ^{208}Tl (β and γ's) both inside of liquid scintillator itself and the balloon lm. In order to remove those backgrounds, we have to not only increase the energy resolution but also use another technique such as Cherenkov ring image.

7. Conclusion

A tetrakis (isopropyl acetoacetato) zirconium and a tetrakis (ethyl acetoacetato) zirconium have a absorption peak at 278 nm and the narrow absorption spectra make less overlapping with the emission of anisole, so that we have succeeded to obtain the liquid scintillator containing those zirconium β-keto ester complex, which has 48.7$\pm$7.1 % relative to BC505 for the light yield and the 4.1$\pm$0.6 % at 3.35 MeV for the energy resolution in case of 40 % photo coverage of ZICOS experiment. Therefore we are almost ready to consider this experiment to be realized. We could estimate the ZICOS experiment should have the sensitivity of $\langle m_\nu \rangle < 0.2 - 0.3$ eV assuming g_A = 1.25, g_{pp} = 1.11 and QRPA model, if the 1.5m radius of balloon will be filled with a liquid scintillator containing 10 wt.% concentration of $Zr(iprac)_4$ which includes 58.5% enriched ^{96}Zr.

[1] K.Abe *et al*. (T2K Collaboration) Phys. Rev. D 88 (2013) 032002.

[2] F.P.An *et al*. Rhys. Rev. Lett. 108 (2012) 171803, J.K.Ahn

et al.(RENO Collaboration) Phys. Rev. Lett. 108 (2102) 191802, Y.Abe et al.(Double Chooz Collaboration) Phys. Rev. Lett. 108 (20122) 131801.

[3] Y.Fukuda et al.(Super-Kamiokande Collaboration) Phys. Rev. Lett. 81 (1998) 1562

[4] S.Fukuda et al. (Super-Kamiokande Collaboration) Phys. Lett. B 539 (2002) 3, Q.R.Ahmed et al. (SNO Collaboration) Phys. Rev. Lett. 89 (2002) 011302, G. Bellini et al. (Borexino Collaboration) Phys. Rev. Lett. 107 (2011) 141302

[5] T.Araki et al. (KamLAND Collaboration) Phys. Rev. Lett. 94 (2005) 081801.

[6] M.Fujii, K.Hamaguchi and T.Yanagida Phys. Rev. D 65 (2002) 115012.

[7] J.Hisano, T.Moroi, K.Tobe, M.Yamaguchi, T.Yanagida Phys. Lett. B 357 (1995) 579

[8] M.Mori, Nucl. Inst. and Meth. A, 732 (2013) 316.

[9] R.Arnold et al.(NEMO-3 collaboration), Phys. Rev. D 89 (2014) 111101.

[10] J. Argyriades (NEMO collaboration), Phys. Rev. C 80 (2009) 032501.

[11] M.B.Kauer Doctor of Philosophy of UCL (2010), J.Argyriades et al. Nucl. Phys. A 847 (2010) 168.

[12] J.Mott Doctor of Philosophy of UCL (2013), R. Arnold et al. (NEMO collaboration), Nucl. Phys. A 765 (2006) 483.

[13] M.Agostini et al. Phys. Rev. Lett 111 (2013) 122503.

[14] P.Belli it et al. Phys. Rev. C 85 (2012) 044610, T.Bloxham et al. (The COBRA collaboration) Phys. Rev. C 76 (2007) 025501.

[15] C.Amaboldi et al. Phys. Rev. C 78 (2008) 035502

[16] J.J. Gomez-Cadenas et al.(The NEXT collaboration), Advances in High Energy Physics, vol. 2014, Article ID 907067, special issue.

[17] M.Auger et al., Phys. Rev. Lett. 109, (2012) 032505, The EXO-200 collaboration Nature 510 (2014) 229.

[18] A.Gando et al. (KamLAND-Zen collaboration), Phys. Rev. Lett.110 (2013) 062502

[19] L.Winslow and R.Simpson, J.INST 7 (2012) P07010.

[20] H.Grubbs et al. Org. Matt. 15 (1996) 1518

[21] C.Back, F.X.Hartmann, D.Motta, S.Schoenert, Chem. Phys.Lett. 435 (2007), 252.

[22] Y. Fukuda, S. Moriyama and I. Ogawa, Nucl. Instr. and Meth. A 732 (2013) 397.

[23] R.Arnold, et al. Nucl. Instr, and Meth. A 563 (2004) 79.