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 tetrakis (isopropyl acetoacetato) zirconium has been developed for new project of neutrinoless double beta decay experiment (ZICOS experiment). We have synthesized a tetrakis (isopropyl acetoacetato) zirconium, which have high solubility (over 31.2 wt.%) in anisole. We measured the performance of liquid scintillator containing 10 wt.% concentration of a tetrakis (isopropyl acetoacetato) zirconium, 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, respectively. We also estimated that ZICOS experiment should be sensitive to $\langle m_\nu \rangle < 0.1$ eV assuming $g_A = 1.25$, $g_{pp} = 1.11$ and QRPA model, if a radius of the inner detector is 1.5 m and the detector is filled with this liquid scintillator with an enriched ^{96}Zr nucleus and we can reduce ^{208}Tl backgrounds to be one tenth order of magnitude of KamLAND-Zen using Cherenkov lights.

1. Introduction

To determine the Majorana neutrino mass from neutrinoless double beta decay ($0\nu\beta\beta$), we must measure the half-life. The half-life of $0\nu\beta\beta$ is given by following formula.

$$[T^{0\nu}_{1/2}(0^+ \to 0^+)]^{-1} = G_{0\nu} M^2_{0\nu} \frac{\langle m_\nu \rangle^2}{m_e^2} \tag{1}$$

Here $G_{0\nu}$ is the kinematic phase space factor, $M_{0\nu}$ is the matrix element of the target nucleus 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 experimentally expressed by following relation.

$$T^{0\nu}_{1/2} \sim a \sqrt{\frac{MT}{\Delta E\, B}} \tag{2}$$

Here 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) experiment for neutrinoless double beta decay search. The spherical detector will be filled with a liquid scintillator which contains a high concentration of tetrakis (isopropyl acetoacetato) zirconium (Zr(iprac)$_4$). If the radius of this detector is 1.5 m, then total volume is 14.1 m^3. This detector is located in an outer cylindrical tank as shown in the left panel of Fig 1, which is 5.4 m in diameter and is 5 m in height, assuming EGADS tank which located in the Kamioka mine [1]. This tank should be filled with a pure water in order to identify the passing muons and to exclude external γs and neutrons. Photomultiplier will be mounted on the wall of both an inner detector and an outer cylindrical tank. The photo coverage of the inner detector should be 40 % in order to collect the scintillation light efficiently.

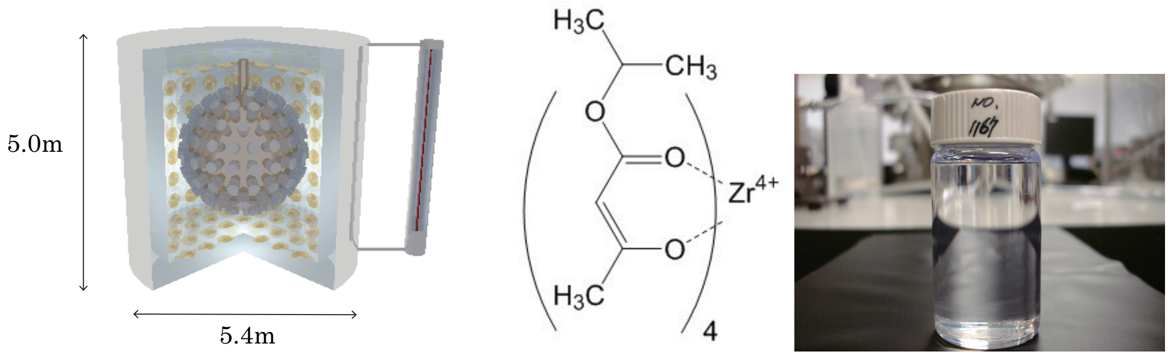

Figure 1. The left panel shows the conceptual design of the ZICOS detector. The spherical inner detector will be filled with a liquid scintillator which contains high concentration of Zr(iprac)$_4$. The middle panels shows the chemical structure formula of Zr(iprac)$_4$ and the right panel shows a sample liquid scintillator containing 10 wt.% concentration of Zr(iprac)$_4$.

A nucleus ^{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. We choose an anisole (methoxybenzene) as a solvent, because it can dissolve Zr(iprac)$_4$ with high concentration. 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. The light yield of the standard cocktail is almost same as BC505 and the decay time is 20 ns.

The complex Zr(iprac)$_4$ is not commercial product, therefore we have to synthesize it by ourselves. The chemical structure formula of Zr(iprac)$_4$ is shown in the middle panel of Fig. 1, and the chemical formula of Zr(iprac)$_4$ is $Zr(CH_3CCOCHCOOCH(CH_3)_2)_4$ (MW=663.87). Synthesized Zr(iprac)$_4$ was a white powder. We measured the solubility of Zr(iprac)$_4$ in anisole and they were over 31.2 wt.%. This corresponds to 70g/L for the solubility of Zr metal. This quite high values makes us to realize tons scale experiment with small size of detector for low backgrounds and low systematic errors. Indeed a quit transparent scintillator cocktail even for the 10 wt.% concentration of Zr(iprac)$_4$ was obtained as shown in the right panel of Fig. 1.

An absorption peak of Zr(iprac)$_4$ was found at 278 nm. The overlap region between the absorption spectrum of Zr(iprac)$_4$ and the emission spectrum of anisole is smaller than the case of Zirconium (IV) Acetylacetonate [2], therefore we could expect good performance of liquid scintillator even if 10 wt.% concentration of Zr(iprac)$_4$ dissolved in anisole.

3. Performance of liquid scintillator

The performance of a liquid scintillator from the point of view of $0\nu\beta\beta$ 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 Zr(iprac)$_4$.

We measured liquid scintillator samples with several concentrations of Zr(iprac)$_4$. The left panel of Fig. 2 shows the measured light yield fraction for the standard cocktail as a function of the concentration of Zr(iprac)$_4$. The light yield could be fitted by using following Eq.(3).

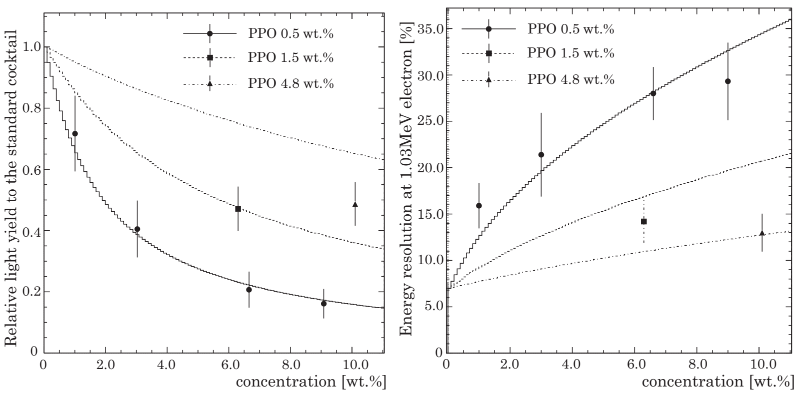

Figure 2. The left panel shows the measured light yield fraction to standard cocktail as a function of the concentration of Zr(iprac)$_4$ in case of the concentration of PPO as 0.5 wt.%, 1.5 wt.% and 4.8 wt.%. The right panel shows the measured energy resolution as a function of the concentration of 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.%

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

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 in mole unit. We assume the emission photon could be absorbed by both PPO and Zr(iprac)$_4$ with a probability in proportion to the number of molecular and the absorbance, because each absorption spectral shape of PPO and Zr(iprac)$_4$ are almost similar. According to the solid line in left panel of Fig. 2 which is fitted by Eq(3), the light yield fraction is expected to be almost 15 % at 10 wt.% concentration of Zr(iprac)$_4$. This number is quite smaller than our goal.

The right panel of Fig. 2 shows the measured energy resolution as a function of the concentration of Zr(iprac)$_4$. 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 function of concentration of Zr(iprac)$_4$. The energy resolution obtained by solid line around 10 wt.% concentration was 35 % at 1.03 MeV. The photo coverage of the measurement setup was estimated to be about 9.3 % using Monte 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 goal. Therefore, we have to improve the liquid scintillator system in order to get both larger light yield and better energy resolution.

According to Eq.(3), the light yield also should be in proportion to fraction of the amount PPO molecular with respect to the amount of Zr(iprac)$_4$ molecular. In other words, we could 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. 2 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 modified by same equation as shown in the right panel of Fig. 2. Actually, if we use the concentration of PPO as 5 wt.% in 10 wt.% concentration of Zr(iprac)$_4$, the energy resolution is estimated by 4.1 ± 0.6 % at 3.35 MeV assuming 40 % photo coverage of photomultiplier. Therefore we achieved our goal for the liquid scintillator system.

4. Sensitivity of ZICOS experiment for $0\nu\beta\beta$ search

An experimental results of $0\nu\beta\beta$ for ^{96}Zr were obtained by NEMO-3 experiment [3]. A lower limits of the life-time was $T^{0\nu}_{1/2} > 9.2 \times 10^{21}$ years and an 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.

ZICOS experiment will use 216 kg of zirconium 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[4]. Assuming same energy resolution, background rates, and the measurement time as those of KamLAND-Zen, we can estimate the sensitivity for the lifetime measurement to be $T^{0\nu}_{1/2} > 4.4 \times 10^{24}$ years for ZICOS experiment. However, this is not enough for $0\nu\beta\beta$ search.

In order to increase sensitivity, we have to use some improvements. One improvement is an enrichment. NEMO-3 experiment used 7g of ^{96}Zr with an enriched to 57.3 % for their target [5]. 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 $T^{0\nu}_{1/2} > 1.1 \times 10^{25}$ years. This is quite 1155 times longer than NEMO-3 limits. Using Eq.(2), it corresponds to $\langle m_\nu \rangle < 0.16 - 0.3$ eV assuming same parameters of $g_A = 1.25$, $g_{pp} = 1.11$ and QRPA model.

Another improvement is to reduce backgrounds around 3.5 MeV region. According to recent analysis of KamLAND-Zen, they found that those backgrounds consist of decay products from ^{208}Tl (β and 2.6 MeV γ) both inside of liquid scintillator and the balloon film. In order to remove those backgrounds, we have to not only increase the energy resolution but also use another technique such as Cherenkov lights.

5. Cherenkov lights using for background reduction

The number of photons generated by the Cherenkov radiation is calculated by following formula.

$$\frac{dN}{dx} = 2\pi z^2 \alpha \sin^2\theta_c \int_{400\mathrm{nm}}^{550\mathrm{nm}} \frac{d\lambda}{\lambda^2} = 277 z^2 \sin^2\theta_c \ \mathrm{photon/cm} \tag{4}$$

A refractive index of anisole is 1.518, so that the light yield of Cherenkov light is 118 photon/cm above 400nm. The Cherenkov light below 400nm should be absorbed by PPO for a re-emission of scintillation. On the other hands, the light yield of scintillation for anisole is almost 12,000 photon/MeV, and the range of 1 MeV electron in anisole is almost 0.8cm, so that the yield ratio of Cherenkov light and scintillation light should be order of 1 %.

In order to confirm the light yield of Cherenkov lights, we used UV cut filter (SC-37) which cuts lights below 400 nm for the measurement of only Cherenkov light. The left panel of Fig. 3 shows the pulse height distribution of no filter, which include not only scintillation lights from anisole (most of them should be less 400 nm) but also Cherenkov lights. The right panel shows only Cherenkov lights above 400 nm using filter. The difference between those amounts corresponds to the light yield of scintillation from anisole, and then the yield ratio is experimentally obtained by 2.8 %. The measured value is almost consistent with the

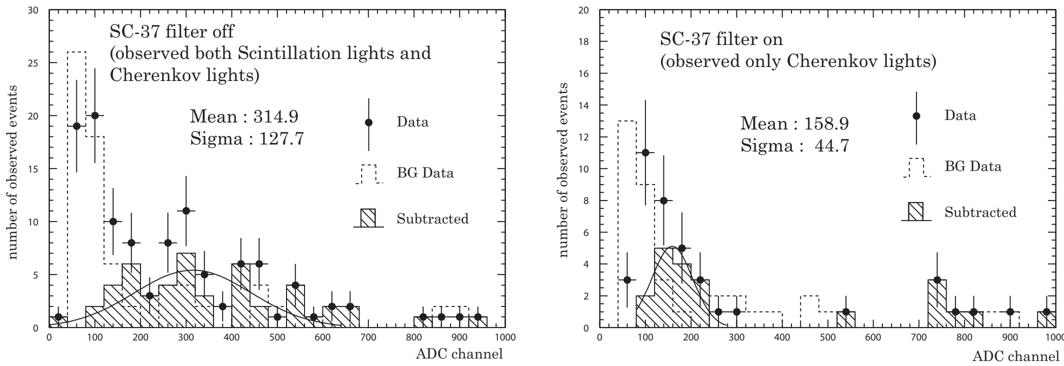

Figure 3. The left panel shows the pulse height distribution of no filter, therefore the light includes both scintillation and Cherenkov. The right panel shows only Cherenkov light distribution above 400 nm, which is obtained by using SC-37 UV cut filter.

expectation within a statistical error. If we can select photomultiplier which detects Cherenkov lights among all hits, then we may be able to distinguish $0\nu\beta\beta$ events and the backgrounds due to the inconsistency of vertexes. For the purpose, we will use the difference of time spread of Cherenkov light (less than 1 ns) and scintillation light (20 ns), which is also argued in Ref.[6].

6. Conclusion

A tetrakis (isopropyl acetoacetato) zirconium has a absorption peak at 278 nm and the narrow absorption spectrum makes less overlapping with the emission spectrum of anisole, so that we have succeeded to obtain that the liquid scintillator containing 10 wt.% concentration of Zr(iprac)$_4$ has 48.7 ± 7.1 % relative to BC505 for the light yield and the 4.1 ± 0.6 % at 3.35 MeV for the energy resolution in case of 40 % photo coverage of photomultiplier. We could estimate the ZICOS experiment should have the sensitivity of $\langle m_\nu \rangle < 0.1$ eV, if we use both 58.5 % enriched ^{96}Zr and a further background reduction method using Cherenkov light in order to achieve one tenth order of magnitude of KamLAND-Zen.

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