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 the Zirconium Complex in Liquid Scintillator (ZICOS) experiment which is a 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 a tetrakis (isopropyl acetoacetato) zirconium, which have high solubility (over 30 wt.%) in anisole. We measured the performance of liquid scintillator containing 10 wt.% concentration, and obtained $48:7:1$% 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, respectively. We also estimated that ZICOS experiment should be sensitive to $\langle m \rangle < 0.1$ 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 0.1 level 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.

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

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.

$$T_{1/2}^{0\nu} \sim a \sqrt{\frac{MT}{\Delta E B}}$$

Here $a$ is the abundance of the target isotope, $M$ is the target mass, $T$ is the measurement time, $\Delta E$ 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 ($\text{Zr(iprac)}_4$). 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[1]. This tank should be filled with a pure water in order to identify the passing muons and to exclude external $\gamma$s 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. To eliminate cosmic muons, the detector should be located in an underground laboratory, such as the Kamioka Observatory.

![Figure 1](image)

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 $\text{Zr(iprac)}_4$. The middle panels shows the chemical structure formula of $\text{Zr(iprac)}_4$ and the right panel shows a sample liquid scintillator containing 10 wt.% concentration of $\text{Zr(iprac)}_4$.

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 zirconium complex 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. This standard cocktail has almost same light yield as BC505 and a fast decay time ($\sim 20$ ns).

A tetrakis (isopropyl acetoacetato) zirconium ($\text{Zr(iprac)}_4$) is not commercial products, therefore we have to synthesize it by ourselves. A chemical structure formula of $\text{Zr(iprac)}_4$ is shown in the middle and middle panel of Fig. 1, and the chemical formula of $\text{Zr(iprac)}_4$ is $\text{Zr(CH}_3\text{CCOCHCOOH(CH}_3\text{)}_2_4$ (MW=663.87). Synthesized $\text{Zr(iprac)}_4$ was a white powder. We measured the solubility of $\text{Zr(iprac)}_4$ 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 detector with small size of detector for low backgrounds and systematic. Indeed a quit transparent scintillator cocktail even for the 10 wt.% concentration of $\text{Zr(iprac)}_4$ was obtained as shown in the right panel of Fig. 1.
An absorption peak of Zr(iprac)$_4$ was found at 278nm. The overlap region between the absorption of Zr(iprac)$_4$ and the emission 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 $\gamma$ 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)$_4$ 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.%

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

Here, $\eta_1$ and $\eta_2$ show the absorbance of PPO and zirconium $\beta$-keto ester complex per mole, and $N_{PPO}$ and $N_{Zr}$ show the amount of PPO and zirconium $\beta$-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 fitted line in Fig. 2 using Eq(3), the light yield fraction to the standard cocktail for Zr(iprac)$_4$ is expected to be almost 15% at a 10 wt.% concentration. 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 $\sigma_0$ 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. 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 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.

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 $T_{1/2}^{0\nu} > 9.2 \times 10^{21}$ year and a upper limit of the neutrino effective mass was also $(m_\nu) < 7.2 - 10.8$ 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 6.5 kg 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 as $T^{0\nu}_{1/2} > 4.4 \times 10^{24}$ years for ZICOS experiment. This is not enough for $0\nu\beta\beta$ experiment.

In order to increase sensitivity, we have to use another 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 $(m_\nu) < 0.16 - 0.3$ eV assuming same parameters of $g_A = 1.25$, $g_{pp} = 1.11$ and QRPA model.

Another improvement is reducing backgrounds around 3.5 MeV region at one order magnitude smaller than KamLAND-Zen. According to recent analysis of KamLAND-Zen, they found that those backgrounds consist of decay product from $^{208}$Tl ($\beta$ and 2.6 MeV $\gamma$) 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.

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

The 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 re-emission of scintillation. The light yield of scintillation for anisole is almost 12,000 photon/MeV, and the range of 1 MeV electron in anisole is almost 0.8 cm, so that the ratio 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 400nm 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 400nm) but also Cherenkov lights. The right panel shows only Cherenkov lights above 400nm using filter. The difference between those amounts corresponds to the light yield of scintillation from anisole, and the ratio between Cherenkov and scintillation is obtained by 3%. The measured value is almost consistent with the expectation.

**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 400nm using filter.

If we can select photomultiplier which detects Cherenkov lights among all hits, then we may be able to distinguish 0νββ events and the backgrounds due to the vertex inconsistency. For that purpose, we will use the difference of time spread (rising and decay time of signals) of the Cherenkov light (less than 1 ns) and the scintillation light (~20 ns).

**6. Conclusion**

A tetrakis (isopropyl acetoacetato) zirconium has an 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 that the liquid scintillator with 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 using a liquid scintillator with 10 wt.% concentration of Zr(iprac)₄ which include 58.5% enriched $^{96}$Zr nucleus and new backgrounds reduction method with Cherenkov light to obtain 0.1 level of KamLAND-Zen.

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**References**