ジルコニウム96を用いたニュートリノを放出しない二重ベータ崩壊事象の探索

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Liquid Scintillator:
(1) 10 wt.% Zr(iprac)_4 loaded in anisole
(2) 3.5% at 3.35MeV of energy resolution with 40% photo coverage and long attenuation length (> 6m) was obtained.

Pure water surrounding inner detector in order to veto muons and external backgrounds.

Inner detector with 40%~60% photo coverage 20” HPD PMT including 1.7ton Zirconium loaded 113 tons LS in fiducial volume. (Total vol. : 180 tons)
Backgrounds around Q-value

Measured by KamLAND-Zen


$0\nu\beta\beta$ signal region for $^{96}$Zr

$T_{1/2}^{0\nu} > 5 \times 10^{25}$y

BG reduction should be necessary.

Lowering BG ($^{208}$Tl) $< 1/20 \times$ KL-Zen ($\sim 1.0$ events/ton/year)

$^{208}$Tl In balloon

(90% C.L. U.L.)
Main backgrounds


Need additional technique other than the energy spectral shape obtained by scintillation lights in order to reduce remaining backgrounds.
Decay mode

\[ ^{208}\text{Tl} : \beta \text{ and } 2.6\text{MeV} \gamma \text{s} \]

\[ ^{214}\text{Bi} : \beta \text{ and multi } \gamma \text{s (609keV, 1.12MeV)} \]

- **Multiple events**
  - \( \beta \) and gammas (2.6MeV + etc) for \(^{208}\text{Tl}\)
  - \( \beta \) and complicated gammas, and \( \alpha \) for \(^{214}\text{Bi}\)

- **Mis-reconstruction and mis-identification**
  - Reconstructed as one event even if multiple events
  - Caused contamination in fiducial volume

\[ ^{208}\text{Tl (3.053 m)} \]

\[ ^{208}\text{Tl} \rightarrow ^{204}\text{Pb} \]

\[ ^{214}\text{Bi} \]

\[ ^{214}\text{Bi} \beta\text{-decay with multi-}\gamma\text{s} \]

\[ ^{214}\text{Bi} \rightarrow ^{214}\text{Po} \]

\[ ^{214}\text{Po} \rightarrow ^{210}\text{Pb} \]

\[ 164\mu s \]
How to distinguish $^{208}$Tl and DBD

- Balloon or surface of detector
- $\beta$ decay
- $2.6\text{MeV} \gamma$
- $0\nu\beta\beta$ event
- Reconstructed vertex by scintillation light
- Reconstructed vertex by Cherenkov light
Simulation of $^{208}\text{Tl}$ decay

1) E : 3.0-3.7MeV 17925 events
2) Fiducial volume 628 events
3) Multi events 263 events
3) Closer events $(d \leq 10\text{cm})$ 35 events

1/20 BG reduction could be achieved.
Light yield of Cherenkov lights

Cherenkov light yield ($\lambda > 400$nm)
Scintillation light yield of anisole

$\approx 0.02 \equiv \approx 200$ photon/MeV
Even though multiple scattering of electrons, Cherenkov light looks have some clusters in the directional distribution.
Angular dependence

Data indicated same angular dependence as MC simulation of Cherenkov lights with EGS5.
Comparison between MC and Data

EGS5 based MC almost reproduced the angular dependence of data.

Cherenkov lights emitted by ~1MeV electron really have a directionality.
Design of ZICOS detector

Detector:
1) 180 tons LS: 10 wt.% Zr(iprac)₄ with PPO/POPOP in anisole
Need 500 of 20” HPD with high QE ~0.4 and Tres (~1ns @ 2pe) = 64% coverage

Expected performance:
1) Energy resolution ~2.8%@3.35MeV
2) T₁/₂(0νββ) > 10²⁶ years, if both 1/20 BG reduction and 50% ⁹⁶Zr enrichment could be achieved.
Average angle distribution

Average direction

\[ e^- = \Sigma d_i \]

Average angle = \[ \frac{\Sigma \theta_i}{N_{\text{hit}}} \]

Average angle with respect to averaged direction for single electron seems to have a peak at 48 degree which is almost same as Cherenkov angle.
Hit pattern of DBD (opposite and half E)

Multi events from DBD tend to have a slightly larger angle than single electron.
Hit pattern of $^{208}$Tl (2.6 MeV $\gamma + \beta + \gamma$)

Multi events from $^{208}$Tl tend to have a smaller angle than DBD or single electron, even vertex obtained by scintillation.

Hit pattern could be used for BG reduction.
Reconstructed vertex resolution

- HPD should have better QE \(~0.4\) and timing resolution \(~1\)ns @ 2pe than HK HPD in order to reconstruct the vertex position with \(\sigma\sim10\)cm.

- Could reconstruct the vertex position using even only hit pattern, however need more smart method to get better resolution.
Further 1/20 reduction of $^{208}\text{Tl}$ (and $^{214}\text{Bi}$) than KLZ could be realized by identifying as multi events.

Simulated Cherenkov lights with EGS5 and could reproduce the angular dependence which is seen by data.

Conceptual design for ZICOS detector with 20” HPD with 64% photo coverage was presented.

Need to develop HPD with higher QE (~40%) and better timing resolution (~1ns @ 2pe) in order to obtain vertex resolution $\sigma \sim 10$ cm.

Need smart methods both to recognize multi events whether DBD or BG and to get a vertex position using hit pattern.
Backgrounds around 3.35MeV

214Bi contaminates within 1m inner volume, but most of events could be removed by Bi-Po tagging. Also 214Bi is not serious BG in terms of signal region for 96Zr. Most serious BG is 208Tl for 96Zr.
Hit pattern of Cherenkov lights

Simulated by EGS5 (kinetic energy 1.65MeV)
For calculation of $2\nu\beta\beta$,

$$\frac{d\omega}{dk_1 dk_2 d\cos \theta} \sim \mathcal{F}(Z, \varepsilon_1) \mathcal{F}(Z, \varepsilon_2) k_1^2 k_2^2 (W_0 - \varepsilon_1 - \varepsilon_2)^5 (1 - \beta_1 \beta_2 \cos \theta)$$

$k_i$, electron momenta
$\varepsilon_i = \sqrt{k_i^2 + m_e^2}$: electron energy
$W_0 = Q + 2m_e$: total release energy
$Q$: Q value $m_e$: electron mass
$\theta$: opening angle $\mathcal{F}$: Fermi func.
$\varepsilon_i$ can generate independently within energy conservation.

For calculation of $0\nu\beta\beta$,

Same calculation but $\varepsilon_i$ can only generate with $\varepsilon_1 + \varepsilon_2 = W_0$. 

2e emit Cherenkov lights
Basic distribution of DBD MC sample

- Two electrons were generated at same vertex position with opposite direction.
- Each energy is 1.65MeV.
- Cherenkov photons were generated at each EGS ausgab subroutine with 200 photons/MeV.
- QE and time resolution were assumed by 0.4 and 1ns@2pe for HPD.
Emission and absorption spectra for solvent and solute in standard cocktail

PPO absorbed most of emission lights from anisole.

Effectively the energy was transferred to the secondary scintillator.
Attenuation length of scintillation light from POPOP (~450nm) was obtained as ~6m.

No problem for radius of ZICOS detector.
How to separate Cherenkov and scintillation

Separation of scintillation and Cherenkov lights in PMT signal using waveform of FADC.

C. Shaomin et al. arXiv:1511.09339

Cherenkov has a faster peak than scintillation.
Natural radiative U/Th decay chain
Electromagnetic isotope separation

@ ORNL
Backscattering method

Single peak could be used even in liquid scintillator.
Cross section of gamma in anisole

Most of few MeV range γs interact with anisole via Compton scattering.

At least, one event could have Cherenkov hits.
No relation between event direction and averaged PMT direction was found.

Clustered PMTs should be made by longest track within multiple scattering.
Light yield of scintillation in anisole

Relative scintillation light yield of anisole is 9.8% to standard cocktail (due to difference of quantum efficiency of PMT)
Thallium-208 radiation branch

- $^{208}\text{Tl}$ (3.053 m)
  - $\beta^-$ decay 100.00%
  - $Q_{\beta^-} = 4.994\text{eV}$

### Radiations

<table>
<thead>
<tr>
<th>$y(i)$</th>
<th>$\text{y(i)}$ (Bq-s)$^{-1}$</th>
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<tbody>
<tr>
<td>beta-5</td>
<td>$2.27 \times 10^{-03}$</td>
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<tr>
<td>beta-8</td>
<td>$3.09 \times 10^{-02}$</td>
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<td>beta-10</td>
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<td>beta-11</td>
<td>$2.45 \times 10^{-01}$</td>
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<td>beta-12</td>
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<td>gamma 4</td>
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<td>gamma 6</td>
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<td>gamma 19</td>
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<td>gamma 25</td>
<td>$9.92 \times 10^{-01}$</td>
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LIGHT YIELD COMPARISON BETWEEN BC505 AND STANDARD COCKTAIL

Light yield of BC505 and our standard cocktail (100mg PPO and 10mg POPOP solved in 20mL anisole) is almost same quality.
Peak values decreased as a function of the concentration of Zr(iprac)₄.

Energy resolutions are also getting worth as a function of the concentration of Zr(iprac)₄.
### Physical Constants of SGC Liquid Scintillators

<table>
<thead>
<tr>
<th>Scintillator</th>
<th>Light Output</th>
<th>Wavelength of Maximum Emission, nm</th>
<th>Decay Constant, ns</th>
<th>H:Ce Ratio</th>
<th>Loading Element</th>
<th>Density</th>
<th>Flash Point °C</th>
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<tr>
<td>BC-505</td>
<td>80</td>
<td>425</td>
<td>2.5</td>
<td>1.331</td>
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<td>BC-509</td>
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<td>BC-521</td>
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<td>4</td>
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<td>Gd (to 1%)</td>
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<td>425</td>
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<td>Enr. $^{11}$B (5%)</td>
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<td>-8</td>
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<tr>
<td>BC-525</td>
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<td>425</td>
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<td>1.56</td>
<td>Gd (to 1%)</td>
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<td>0.87</td>
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<td>BC-537</td>
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<td>0.99 (D:C)</td>
<td>$^3$H</td>
<td>0.954</td>
<td>-11</td>
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</table>

*Anthracene light output – 40–50% of NaI(Tl)  

1 Fast component; mean decay times of first 3 components – 3.16, 32.3 and 270 ns

LY of NaI(Tl) : $4 \times 10^4$ photon/MeV  
LY of BC505 : $1.2 \times 10^4$ photon/MeV
RECOVERY FOR ABILITY OF LIGHT YIELD AND ENERGY RESOLUTION

PPO helps recovering the light yield and the energy resolution.

Confirmed our assumption and obtained optimized real cocktail (PPO 5wt.% POPOP 0.5wt.%)
UV sharp cut filter (Fuji films)
Photo coverage
Light yield and energy resolution

$\text{Zr(ipcac)}_4$ and $\text{Zr(etac)}_4$ have almost same performance, but LY and Eres depend strongly on concentration.
Attenuation length of light from POPOP was obtained as ~6m for current liquid scintillator. Attenuation length will be recovered ~15m by same purification method as RENO with Al$_2$O$_3$. (Ref: H.Grubbs et al., Org.Mat. 1996 15, 1518-1520)
Future $0\nu\beta\beta$ experiments

$\sum_i U_{ei}^2 m_i$ \[ m_{\beta\beta} = | \sum_i U_{ei}^2 m_i | \]

IH ($\Delta m^2 < 0$)

NH ($\Delta m^2 > 0$)

$10^{27} - 10^{28}$ y

$~$ tons of target and $~$ zero BG detector will be necessary for next generation $0\nu\beta\beta$ experiment.

Neutrinoless double beta decay

\[ [T_{1/2}^{0\nu}(0^+ -> 0^+)]^{-1} = G_{0\nu}(E_0, Z) |M_{0\nu}|^2 <m_\nu^2>/m_e^2 \]

\[ T_{1/2} \sim a \left( M_t / \Delta E \cdot B \right)^{1/2} \]

- \( a \): abundance
- \( M \): target mass
- \( t \): measuring time
- \( \Delta E \): energy resolution
- \( B \): BG rate

**Requirement:** Low BG, Large target mass, High energy resolution
Neutrino mass sensitivity of ZICOS experiment

Results from NEMO-3 ($^{96}\text{Zr}$): $T_{1/2}^{0\nu} > 9.2 \times 10^{21}\text{y}$

$\langle m_\nu \rangle > 7.2 – 10.8$ eV ($g_A=1.25, g_{pp}=1.11, \text{QRPA}$)

(Ref: M.B.Kauer Doctor thesis for UCL(2010))

Assuming same energy resolution, BG rate and measurement time as KamLAND-Zen

($T_{1/2}^{0\nu} > 2.6 \times 10^{25}\text{y}$)  


Mass: 113 ton $\rightarrow$ 10wt.% Zr(iprac)$_4$ = 12.6ton

includes 1.7ton of Zirconium

= 45 kg of $^{96}\text{Zr}$ (natural abundance 2.6%)

( 64kg of $^{136}\text{Xe}$ = 0.2 × KL-Zen)

$T_{1/2}^{0\nu} > 1.2 \times 10^{25}\text{y}$ ← Not enough for $0\nu\beta\beta$ search
Neutrino mass sensitivity of ZICOS experiment

1) Zr enrichment
58.5% enrichment of $^{96}$Zr (e.g. 57.3% for NEMO-3)
then $^{96}$Zr will be 1.0 ton (4.4 times $^{136}$Xe 320kg)

$$T_{1/2}^{0\nu} > 5 \times 10^{25} \text{y} ; <m_\nu> < 0.09 - 0.15 \text{ eV (QRPA)}$$

2) Lowering BG ($^{208}$Tl /$^{214}$Bi)
i.e. $< 1/20 \times \text{KL-Zen}$
($\sim 1.0\text{events/ton/year}$)

$$T_{1/2}^{0\nu} > 5 \times 10^{25} \text{y}$$

See this talk
Development of Zr loaded LS

Zr(CH$_3$COCHCOOCH(CH$_3$)$_2$)$_4$ : Zr(iprac)$_4$

tetrakis (isopropyl acetoacetate) zirconium

mw : 663.87

Solid crystal or powder
Absorbance spectra for \( \text{Zr(iprac)}_4 \)

Absorption peaks of \( \text{Zr(iprac)}_4 \) was found around at 278nm. However, overlapped region with emission of anisole was existed.

\( \text{Zr(iprac)}_4 \) works as a quencher for the liquid scintillator system.
Solubility of $\text{Zr}(\text{i}p\text{rac})_4$ for anisole

Solubility $> 31.2$ wt.\%

$> 70\text{g/L}$ of Zirconium could be solved in anisole.

$\text{Zr}(\text{i}p\text{rac})_4 2242\text{mg}$, PPO $999\text{mg}$ and POPOP $10\text{mg}$ solved in $20\text{mL}$ Anisole
Light yield = \( L_0 \times \frac{\sigma_1 N_{ppo}}{\sigma_1 N_{ppo} + \sigma_2 N_{Zr}} \)

\( L_0 \) : Light yield of anisole  \\
\( N_{ppo} \) : Number of PPO molecular in mole  \\
\( N_{Zr} \) : Number Zr complex molecular in mole  \\
\( \sigma_1 \) : absorbance of PPO \( (\text{mol}^{-1}) \)  \\
\( \sigma_2 \) : absorbance of Zr complex \( (\text{mol}^{-1}) \)

PPO would help the recovering light yield.
Recovering the light yield

Measured at several conditions of PPO concentration

5wt.% PPO helps actually recovering the scintillation light yield.

48.7 ± 7.1% light yield to standard cocktail was obtained at 10wt.% concentration.
Recovering the energy resolution

Measured at several conditions of PPO concentration

5wt.% PPO helps again the energy resolution
35% $\rightarrow$ 13%. at 10wt.% of Zr(iprac)$_4$.

$$\frac{13.0 \pm 2.0\%}{\sqrt{(40\%/9.2\%) \times (3.35\text{MeV}/1.03\text{MeV})}} = 3.5 \pm 0.5\% \text{ at } 3.35\text{MeV}$$

Achieved goal!
Stability of liquid scintillator


Keep transparent liquid and no precipitate is found.
Measurement of backgrounds from LS

Using subtracted # of events around 2.6MeV and 2.2MeV

$^{214}\text{Bi} < 4.9 \times 10^{-20} \text{g/g}$  $^{208}\text{Tl} < 2.7 \times 10^{-22} \text{g/g}$

$^{238}\text{U} < 6.4 \times 10^{-6} \text{g/g}$  $^{232}\text{Th} < 7.4 \times 10^{-7} \text{g/g}$ (c.f. KL $10^{-18} \text{g/g}$)
Property of Cherenkov light

- Refractive index of anisole: \( n = 1.518 \)
- Cherenkov angle is determined by \( \cos \theta = \frac{1}{n'} \beta \) (\( E_e > 0.7 \text{MeV} \)) \( n' > n \)
- Assuming 1.65 MeV electron, then \( \beta = 0.972 \) and Cherenkov angle \( \theta = 47.3 \) degree are expected.
- Number of Cherenkov photon: 100 photon/MeV (400nm – 600nm)

\[
\frac{dN}{dx} = 2 \pi z^2 \alpha \sin^2 \theta_c \int_{\lambda_1}^{\lambda_2} \frac{d\lambda}{\lambda} = 475 z^2 \sin^2 \theta_c \text{photon/cm}
\]

c.f. Light yield of Scintillation: \(~12000\text{photon/MeV}\)

Cherenkov light = \(~1\%\) of scintillation light
Simulation of $^{208}$Tl decay

1) $E : 3.0$-$3.7$MeV
   1555 events
2) fiducial volume
   91 events
3) Multi events
   22 events
4) Closer events
   5 events

1/20 BG reduction could be achieved