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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Neutrinoless double beta decay

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

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

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

**Requirement:** Low BG, Large target mass, High energy resolution
Zirconium Complex in Organic Liquid Scintillator for neutrinoless double beta decay (ZICOS) experiment

Goals for development of LS:
1. > 10wt.% solubility
2. 3.5% at 3.35MeV of energy resolution, if ZICOS have PMTs with 40% photo coverage and long attenuation length (~15m)

LS surrounding inner balloon to veto external BG

Zirconium loaded 113 tons LS
Goals for development of LS:
1. > 10wt.% solubility
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Zirconium Complex in Organic Liquid Scintillator for neutrinoless double beta decay (ZICOS) experiment

LS surrounding inner balloon to veto external BG

Zirconium loaded 113 tons LS

PMT with 40% photo coverage

inner balloon

3m

13m
Zirconium $\beta$-keto ester complex

$$Zr(CH_3COCHCOOCH(CH_3)_2)_4 = Zr(\text{iprac})_4$$
$$\text{mw} : 663.87$$

$$Zr(CH_3COCHCOOCH_2CH_3)_4 = Zr(\text{etac})_4$$
$$\text{mw} : 607.76$$

Absorption wavelength could be shorten.

Synthesized by Prof. T. Gunji
Solubility of Zirconium $\beta$-keto ester complex for anisole

$\text{Zr(iprac)}_4 : 31.2\text{wt.\%}$  $\text{Zr(etac)}_4 : 32.7\text{wt.\%}$

$> 70\text{g/L of Zirconium could be solved in anisole}$
Absorbance spectra for zirconium β-keto ester complex

Absorption peaks of Zr(iprac)$_4$ was found around at 278nm. Peak moved only 10nm.

Overlap region for the absorption spectrum of Zr(iprac)$_4$ became smaller than that of Zr(acac)$_4$. 

Absorbance spectra for zirconium β-keto ester complex
Light yield calculated by quenching

\[
\text{Light yield} = L_0 \times \frac{\sigma_1 N_{\text{ppo}}}{\sigma_1 N_{\text{ppo}} + \sigma_2 N_{\text{Zr}}}
\]

- **L₀**: Light yield of anisole
- **N_{\text{ppo}}**: Number of PPO molecular in mole
- **N_{\text{Zr}}**: Number Zr complex molecular in mole
- **σ₁**: absorbance of PPO (mol⁻¹)
- **σ₂**: absorbance of Zr complex (mol⁻¹)
Light yield and energy resolution

Zr(ipcac)$_4$ and Zr(etac)$_4$ are almost same performance.
Light yield calculated by quenching

\[
L_0 \times \frac{\sigma_1 N_{\text{ppo}}}{\sigma_1 N_{\text{ppo}} + \sigma_2 N_{\text{Zr}}}
\]

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

\(N_{\text{ppo}}\) should help recovering the light yield.
Modification of light yield

$\text{Zr}(\text{iprac})_4$ in several conditions of PPO concentration.

Light yield decreased as calculated formula. However, PPO helps actually the light yield recovering.

$48.7 \pm 7.1\%$ light yield to standard cocktail was obtained at 10wt.% concentration.
Modification of energy resolution

Zr(iprac)$_4$ in several conditions of PPO concentration

PPO helps again the energy resolution at 10wt.% concentration to be 35% $\rightarrow$ 13%.

$13.0 \pm 2.0\%$

$\sqrt{40\%/9\%} \times 0.72 \times 3.35\text{MeV}/1.03\text{MeV}$

$= 4.1 \pm 0.6\%$ at 3.35MeV

Achieved our initial goal!
Neutrino mass sensitivity of ZICOS experiment

Results from NEMO-3: $T_{1/2}^{0\nu} > 9.2 \times 10^{21}$y  
$\langle m_\nu \rangle > 7.2 - 10.8$ eV ($g_A=1.25, g_{pp}=1.11$, QRPA)

Assuming 3m radius, and same Eres, BG rate and mes.time as KamLAND-Zen ($T_{1/2}^{0\nu} > 1.9 \times 10^{25}$y )

Volume: 113ton $\rightarrow$ 10wt.% Zr(iprac)$_4$ = 12.6ton includes 1.73ton of Zr includes 51.9kg of $^{96}$Zr (0.23 times $^{136}$Xe 320kg)

Sensitivity of ZICOS experiment:
$T_{1/2}^{0\nu} > 4.4 \times 10^{24}$y; $\langle m_\nu \rangle < 0.3 - 0.5$eV ($g_A=1.25, g_{pp}=1.11$, QRPA)

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

To reach for $<m_\nu> < 0.1\text{eV}$

1) If a radius of balloon is 3m,
   - 30wt.% $\text{Zr(iprac)}_4 = 156\text{kg of }^{96}\text{Zr}$
   - $^{96}\text{Zr}$ enriched to 10% - 15% using Centrifuge

   $<m_\nu> < 0.09 - 0.11 \text{eV}$

2) To reach for $<m_\nu> \sim 0.03\text{eV}$
   - need 5tons of $^{96}\text{Zr}$
   - 6m radius + 30wt.% $\text{Zr(iprac)}_4 + ^{96}\text{Zr}$ enriched to 10% - 15%

   Need KamLAND balloon or SNO+ acrylic vessel
SUMMARY

- Liquid scintillator containing zirconium β-keto ester complex for ZICOS experiment was developed.
- The absorption peak of zirconium β-keto ester complex stayed at 268nm, but smaller overlap with emission of anisole than zirconium acetylacetone.
- Liquid scintillator with 10 wt.% concentration of Zr(iprac)_4 has 48.7±7.1% for light yield to BC505 and 4.7±0.8% at 2.5MeV (assuming 40% photo coverage and 15m attenuation length) for energy resolution, so that we have really achieved our initial goal!
- Sensitivity of ZICOS experiment: T_1/2^{0ν} > 4.4 \times 10^{24} y; <m_ν> < 0.3 – 0.5eV (g_A=1.25, g_{pp}=1.11, QRPA) assuming BG rate of KamLAND-Zen.
Synthesis of Tetrakis(diethyl malonato) Zirconium

\[
\text{Zr(CH}_3\text{CH}_2\text{OCOCHCOOCH}_2\text{CH}_3\text{)}_4 = \text{Zr(deml)}_4 \text{ mw : 727.87}
\]

Yellow crystal
Absorbance of Tetrakis (diethyl malonato) Zirconium

peak wavelength : 265.2nm
Zr(deml)$_4$ has a little better performance than Zr(iprac)$_4$. 
BACKUP
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 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)
Attenuation length of light from POPOP was obtained as \(~6m\). It is almost equivalent with the detector size.
Absorbance spectra in hexane / diethyl ether

~280nm peak disappeared and a precipitate appeared.

Most of solved Zr atom was found in the precipitate, and no Zr atom in the residual solvent by ICP - Atomic Emission Spectrometry.
Absorbance spectra in acetonitrile / 2-propanol

They are stable and quite transparent.

Solved Zr atoms were found in every region.

~280nm peak should be due to Zr(iprac)$_4$ and Zr(etac)$_4$. 
## Results from ICP Atomic Emission Spectrometry

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<tr>
<th>搬入日</th>
<th>試料名</th>
<th>ジルコニウム濃度 (mg/L)</th>
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<tr>
<td>平成26年11月28日</td>
<td>ジェチルエーテル Zr(iprac)_4 8.3mg/20mL 上澄み液 2014.11.11 No.1075</td>
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<td>2014.10.17 Zr(iprac)_4 5.9mg ジェチルエーテル No.980</td>
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<td>平成26年12月10日</td>
<td>プロパノール Zr(iprac)_4 (1.5/20) 上5mL</td>
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<td>2-プロパノール(20mL)+Zr(iprac)_4 (1.5mg) 2013.11.22 No.497</td>
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<td>2-プロパノール+Zr(iprac)_4 吸収1回目測定 2013.11.22 No.497から No.525</td>
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<td>アニソール Zr(iprac)_4 2.0/20mL No.673(上)5mL</td>
<td>2.6</td>
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Light yield of BC505 and our standard cocktail (100mg PPO and 10mg POPOP solved in 20mL anisole) is almost same quality.
ENERGY SPECTRA FOR SEVERAL CONCENTRATION OF ZR(iprac)₄

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.
ENERGY SPECTRA FOR SEVERAL CONCENTRATION OF ZR(ETAC)₄

Peak values decreased as a function of the concentration of Zr(etac)₄.

Energy resolutions are also getting worth as a function of the concentration of Zr(etac)₄.
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.%).
Single peak could be used even in liquid scintillator.
Photo coverage

- Generated point in the vial
- Detected scintillation light on PMT surface
- 9.3% of photon was detected by PMT

Light guide

PMT

Liquid scintillator

61
50
15
35

48

Japanese: 日本物理学会 第70回年次大会

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Photon attenuation in ZICOS detector

Assuming 3m radius balloon and 5m radius of tank with 15m attenuation length of anisole, the photon attenuation will be 0.72 in average.
Solubility of PPO in anisole with 10wt.% concentration of Zr(iprac)$_4$ 

\[
\frac{205.5\text{mg}}{205.5\text{mg} + 2.0\text{g}} = 9.3\text{ wt.%}
\]

Maybe solved in 20 wt.% of Zr(iprac)$_4$
high energy resolution
4%@2.5MeV

~tons of target will be necessary for next generation detector

http://kds.kek.jp/getFile.py/access?contribId=37&sessionId=16&resId=2&materialId=slides&confId=9151