

Supported by Grant-in-Aid for Scientific Research (C) 24540295 and  
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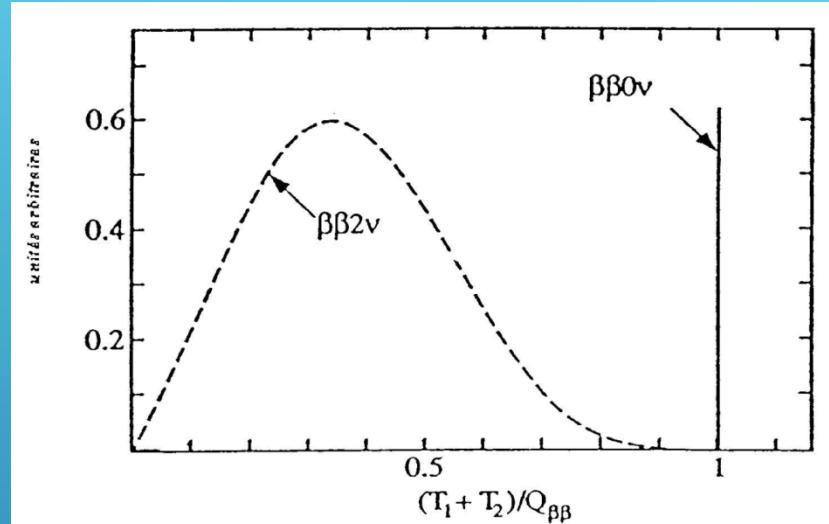
# ジルコニウム96を用いたニュートリノを 放出しない二重ベータ崩壊事象の探索VII

日本物理学会 第70回年次大会  
2015年3月21日

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東京大学宇宙線研究所 森山 茂栄  
福井大学工学部 小川 泉  
東京理科大学理工学部 郡司天博、塚田 学、速水良平

# Neutrinoless double beta decay

$\beta\beta$ emitters with $Q_{\beta\beta} > 2$ Mev		
Transition	$Q_{\beta\beta}$ (keV)	Abundance (%) ( $^{232}Th = 100$ )
$^{110}Pd \rightarrow ^{110}Cd$	2013	12
$^{76}Ge \rightarrow ^{76}Se$	2040	8
$^{124}Sn \rightarrow ^{124}Te$	2288	6
$^{136}Xe \rightarrow ^{136}Ba$	2479	9
$^{130}Te \rightarrow ^{130}Xe$	2533	34
$^{116}Cd \rightarrow ^{116}Sn$	2802	7
$^{82}Se \rightarrow ^{82}Kr$	2995	9
$^{100}Mo \rightarrow ^{100}Ru$	3034	10
$^{96}Zr \rightarrow ^{96}Mo$	3350	3
$^{150}Nd \rightarrow ^{150}Sm$	3667	6
$^{48}Ca \rightarrow ^{48}Ti$	4271	0.2



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

$T_{1/2} \sim a(Mt/\Delta E \cdot B)$       a: abundance      M: target mass

t: measuring time     $\Delta E$ : energy resolution    B: BG rate

Requirement : Low BG, Large target mass, High energy resolution

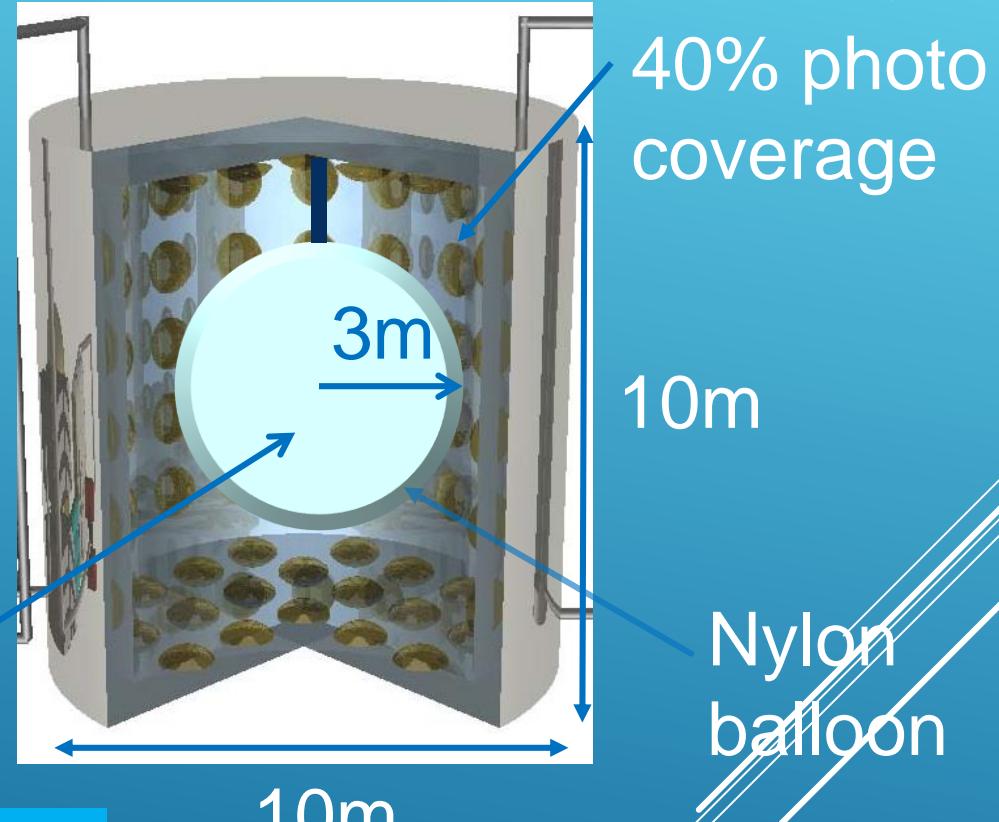
# Zirconium Complex in Organic Liquid Scintillator for neutrinoless double beta decay (ZICOS) experiment

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



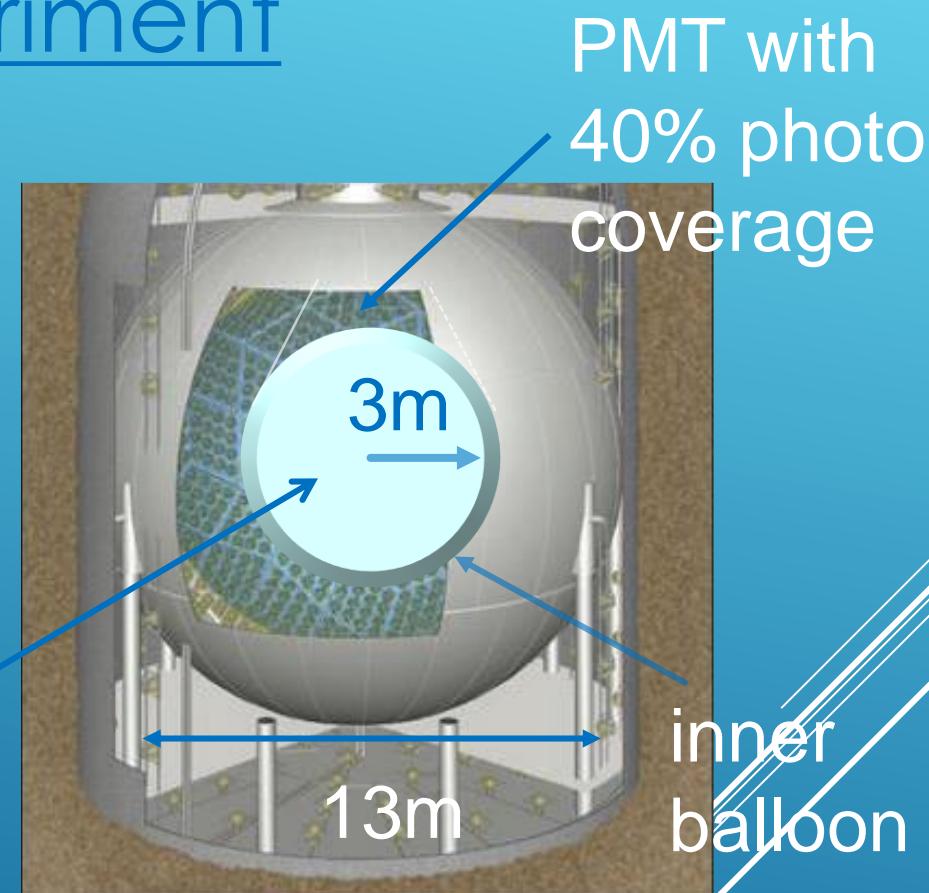
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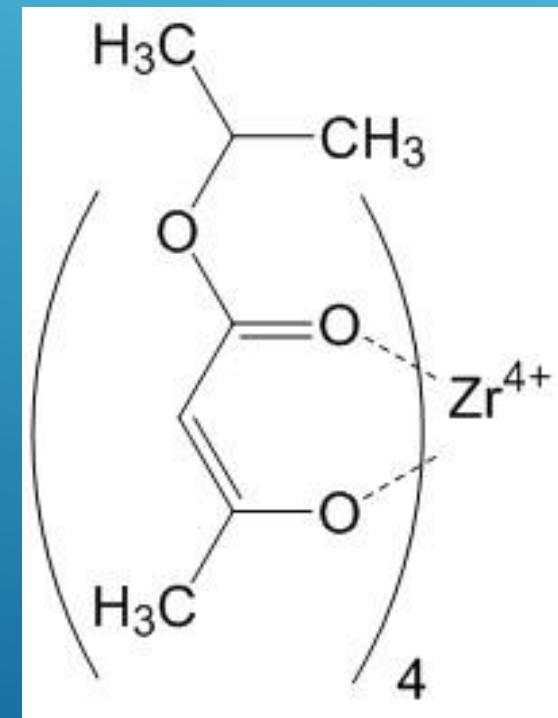


# Zirconium $\beta$ -keto ester complex



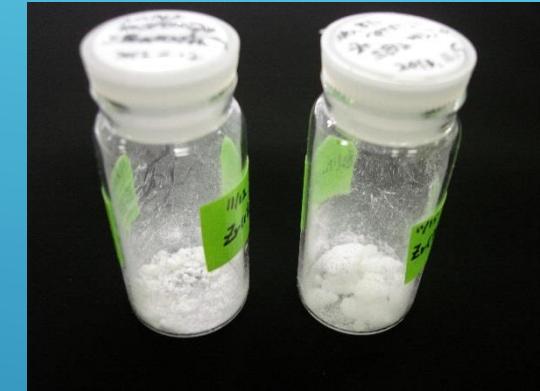
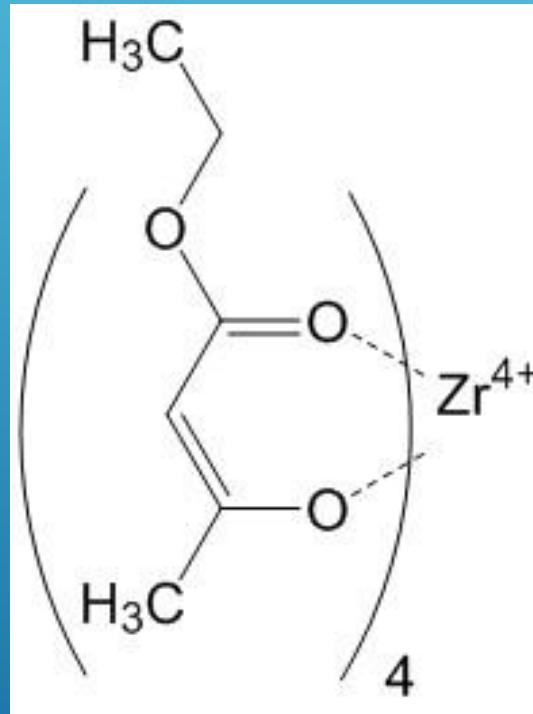
= Zr(iprac)<sub>4</sub>

mw : 663.87



= Zr(etac)<sub>4</sub>

mw : 607.76



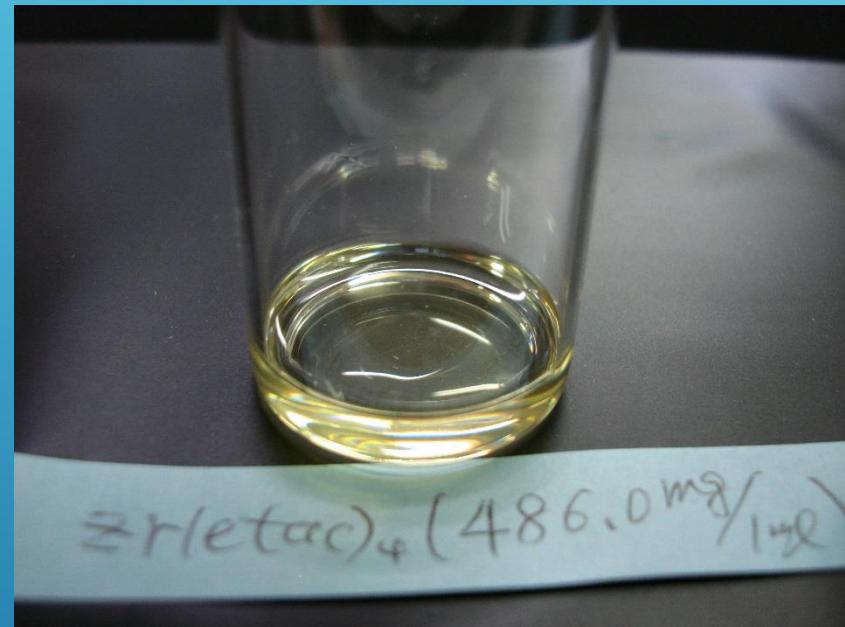
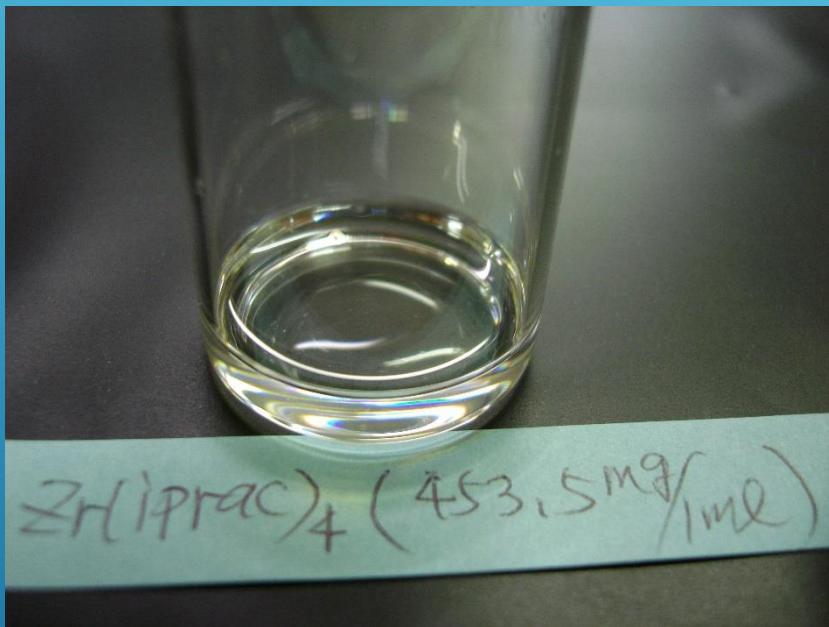
Synthesized by  
Prof. T.Gunji

Absorption wavelength could be shorten.

# Solubility of Zirconium $\beta$ -keto ester complex for anisole

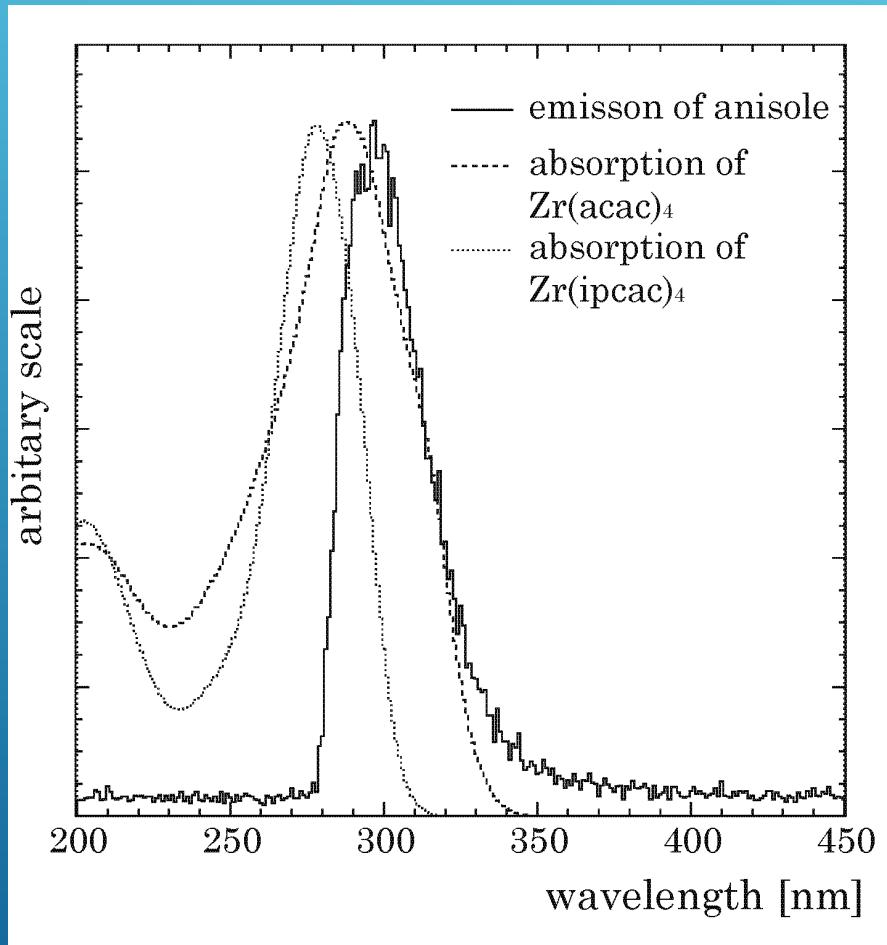
$Zr(iprac)_4$  : 31.2wt.%

$Zr(etac)_4$  : 32.7wt.%



> 70g/L of Zirconium could be solved in anisole

# Absorbance spectra for zirconium $\beta$ -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  $Zr(acac)_4$ .

# Light yield calculated by quenching

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



$L_0$  : Light yield of anisole

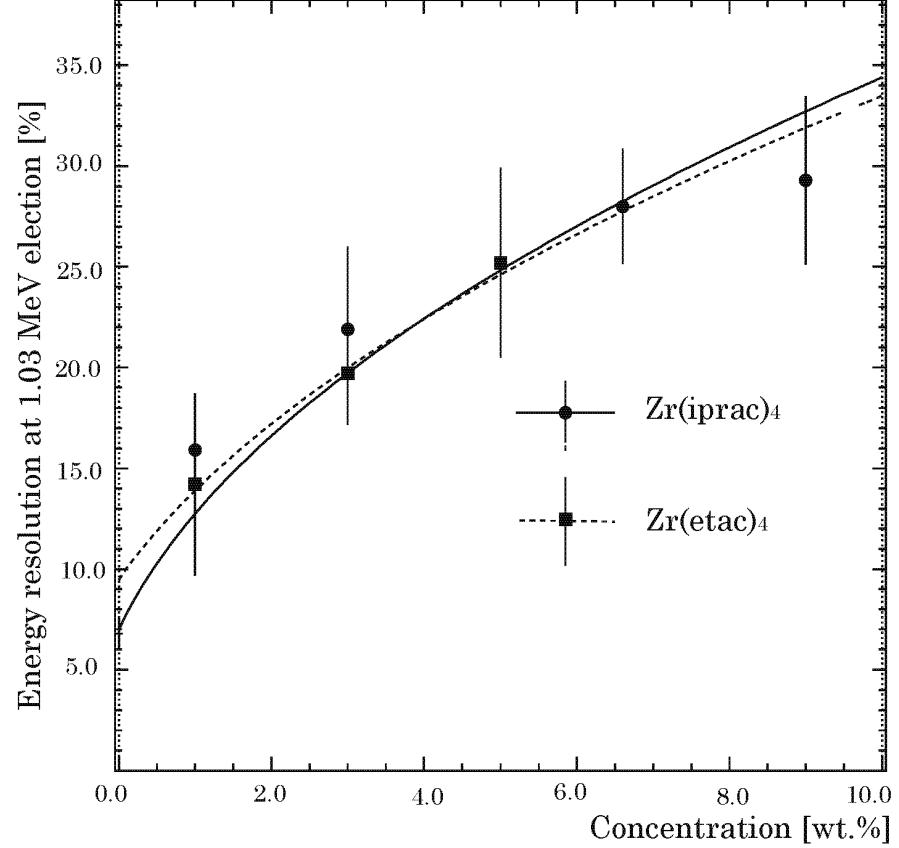
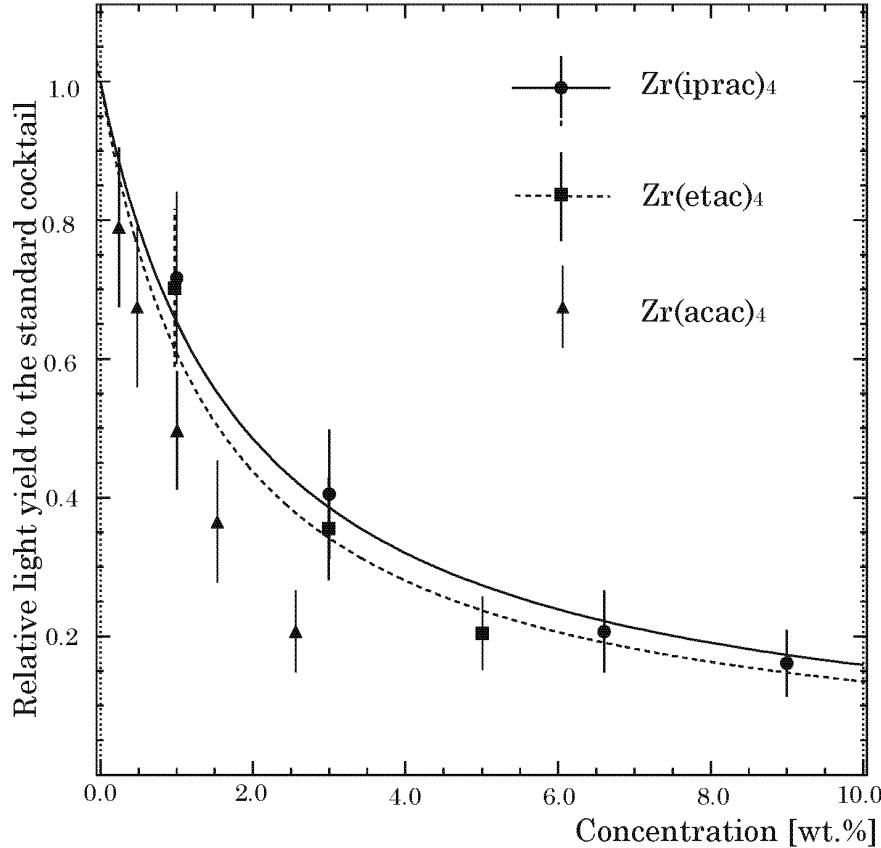
$N_{\text{ppo}}$  : Number of PPO molecular in mole

$N_{\text{Zr}}$  : Number Zr complex molecular in mole

$\sigma_1$  : absorbance of PPO ( $\text{mol}^{-1}$ )

$\sigma_2$  : absorbance of Zr complex ( $\text{mol}^{-1}$ )

# Light yield and energy resolution



Zr(iprac)<sub>4</sub> and Zr(etac)<sub>4</sub> are almost same performance.

# Light yield calculated by quenching

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



$L_0$  : Light yield of anisole

$N_{\text{ppo}}$  : Number of PPO molecular in mole

$N_{\text{Zr}}$  : Number Zr complex molecular in mole

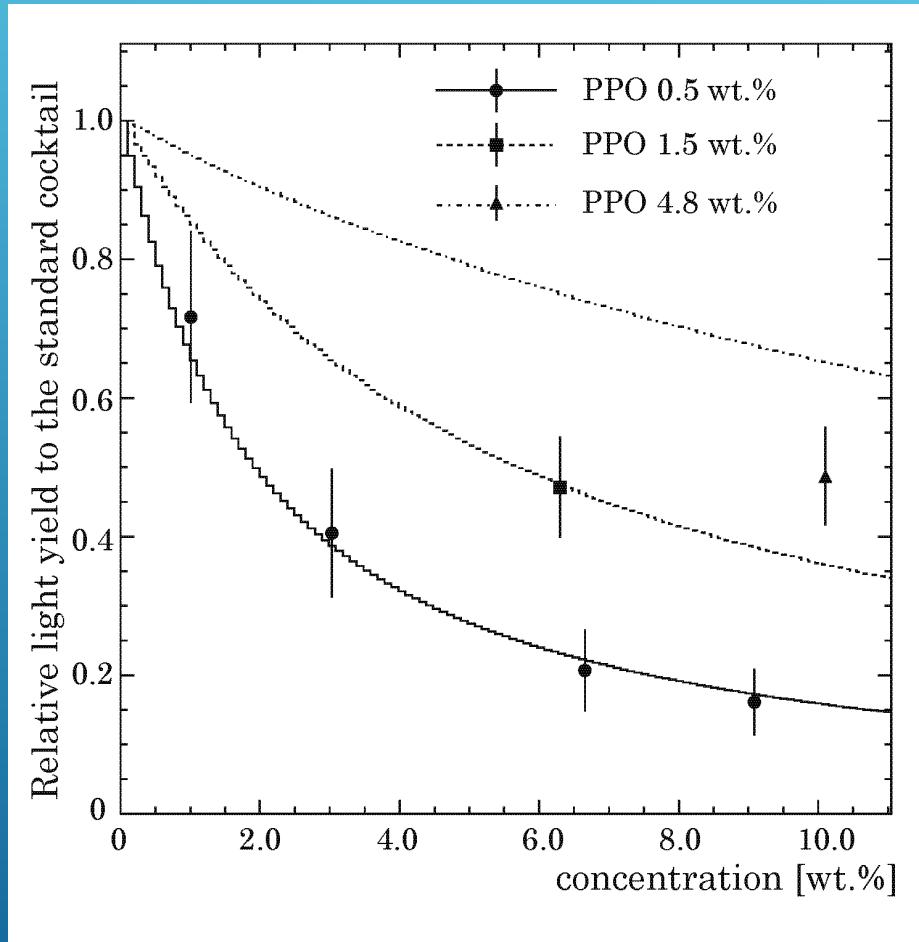
$\sigma_1$  : absorbance of PPO ( $\text{mol}^{-1}$ )

$\sigma_2$  : absorbance of Zr complex ( $\text{mol}^{-1}$ )

**$N_{\text{ppo}}$  should help recovering the light yield.**

# Modification of light yield

Zr(iprac)<sub>4</sub> in several conditions of PPO concentration.



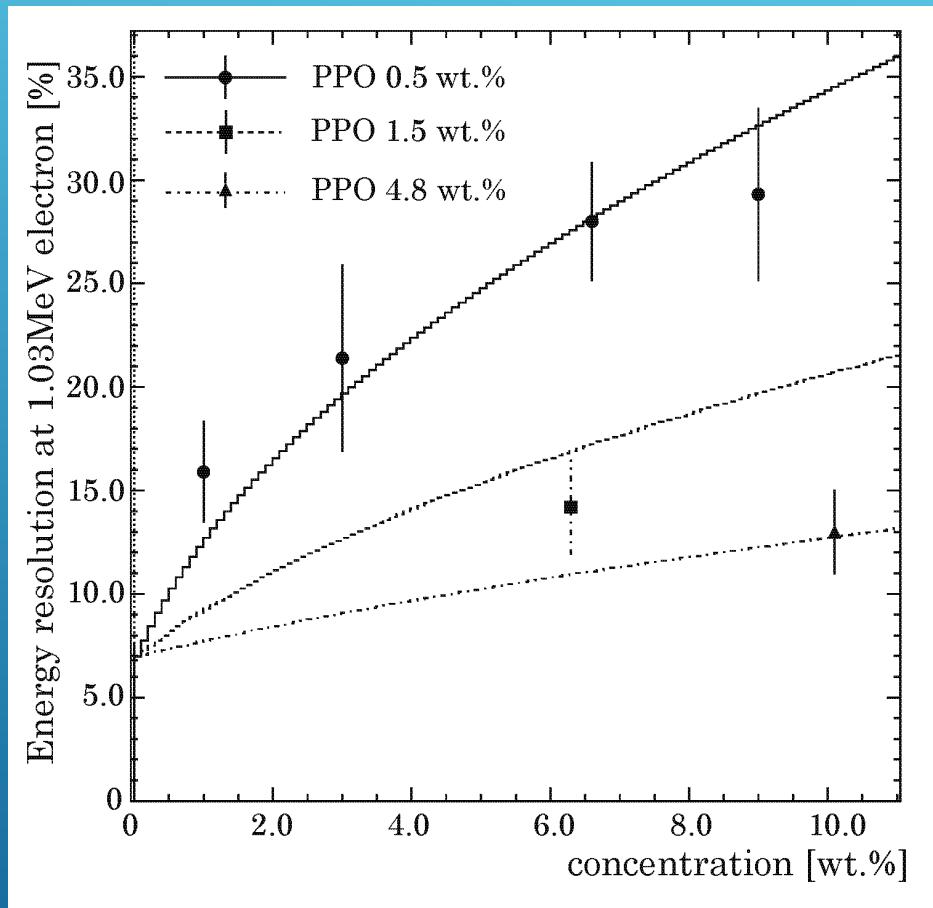
Light yield decreased as calculated formula. However, PPO helps actually the light yield recovering.



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

# Modification of energy resolution

Zr(iprac)<sub>4</sub> in several conditions of PPO concentration



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

$$13.0 \pm 2.0\%$$
$$\sqrt{40\%/9\% * 0.72 * 3.35 \text{MeV} / 1.03 \text{MeV}}$$

$$= 4.1 \pm 0.6\% \text{ at } 3.35 \text{MeV}$$

Achieved our initial goal!

# Neutrino mass sensitivity of ZICOS experiment

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

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

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

(Ref: I.Shimizu arXiv:1409.0077 (2014))

Volume: 113ton  10wt.% Zr(iprac)<sub>4</sub> = 12.6ton  
includes 1.73ton of Zr  
includes 51.9kg of <sup>96</sup>Zr (0.23 times <sup>136</sup>Xe 320kg)

## Sensitivity of ZICOS experiment :

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

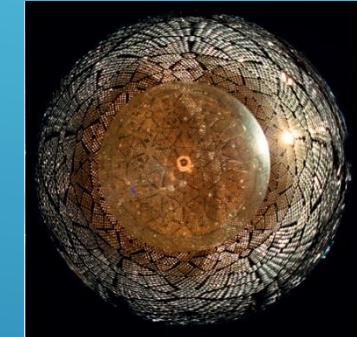
# To reach for $\langle m_\nu \rangle < 0.1\text{eV}$

1) If a radius of balloon is 3m,

- 30wt.% Zr(iprac)<sub>4</sub> = 156kg of <sup>96</sup>Zr
- <sup>96</sup>Zr enriched to 10% - 15% using Centrifuge



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



2) To reach for  $\langle m_\nu \rangle \sim 0.03\text{eV}$

- need 5tons of <sup>96</sup>Zr
- 6m radius + 30wt.% Zr(iprac)<sub>4</sub> + <sup>96</sup>Zr enriched to 10% - 15%



Need KamLAND balloon or SNO+ acrylic vessel

# SUMMARY

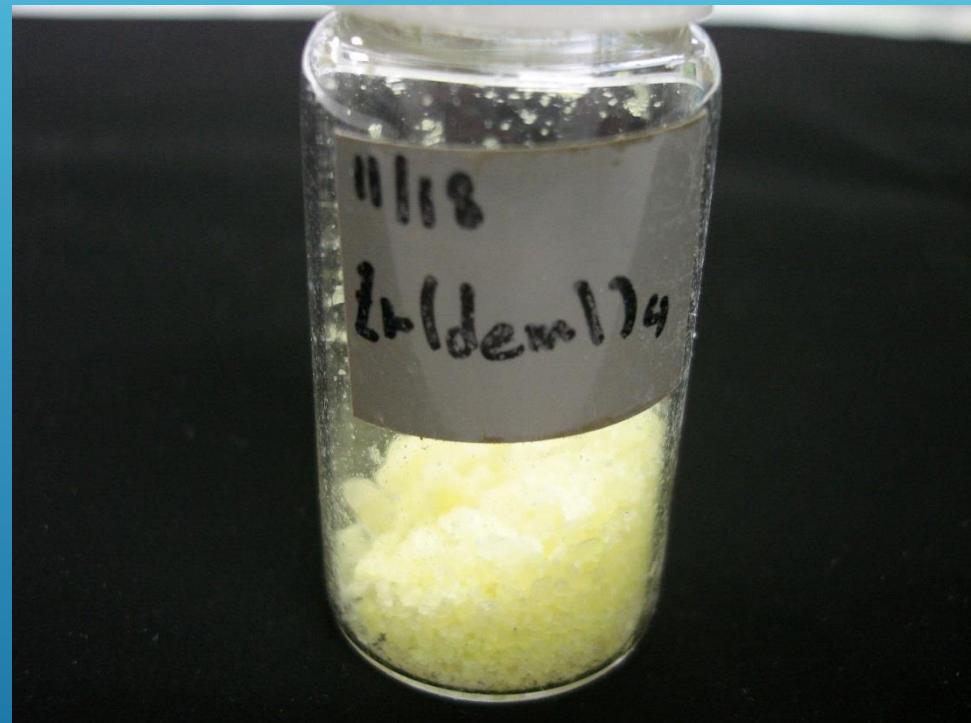
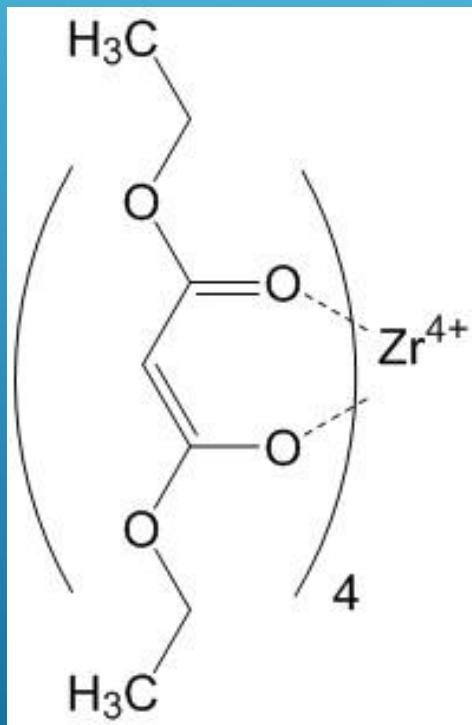
- ▶ Liquid scintillator containing zirconium β-keto ester complex for ZICOS experiment was developed.
- ▶ The absorption peak of zirconium β-keto ester complex stayed at 268nm, but smaller overlap with emission of anisole than zirconium acetylacetone.
- ▶ Liquid scintillator with 10 wt.% concentration of  $\text{Zr(iprac)}_4$  has  $48.7 \pm 7.1\%$  for light yield to BC505 and  $4.7 \pm 0.8\%$  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 :  $T_{1/2}^{0\nu} > 4.4 \times 10^{24}\text{y}$ ;  $\langle m_\nu \rangle < 0.3 - 0.5\text{eV}$  ( $g_A = 1.25, g_{pp} = 1.11$ , QRPA) assuming BG rate of KamLAND-Zen.

# Synthesis of Tetrakis(diethyl malonato) Zirconium

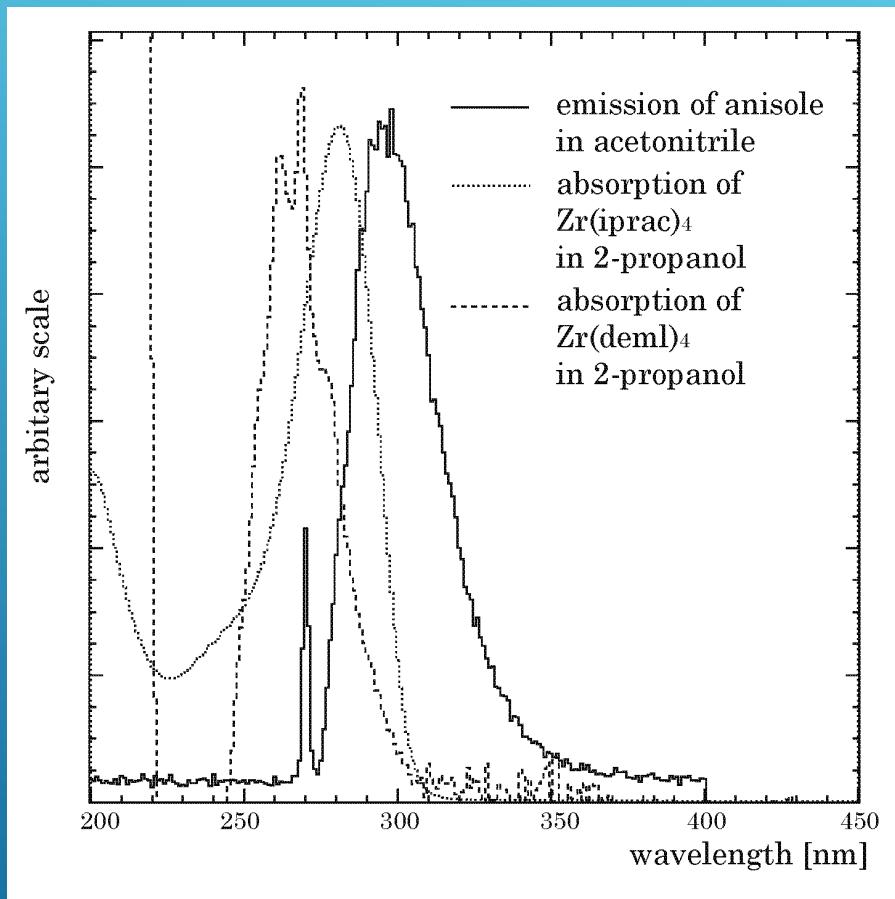


=  $\text{Zr}(\text{deml})_4$  mw : 727.87

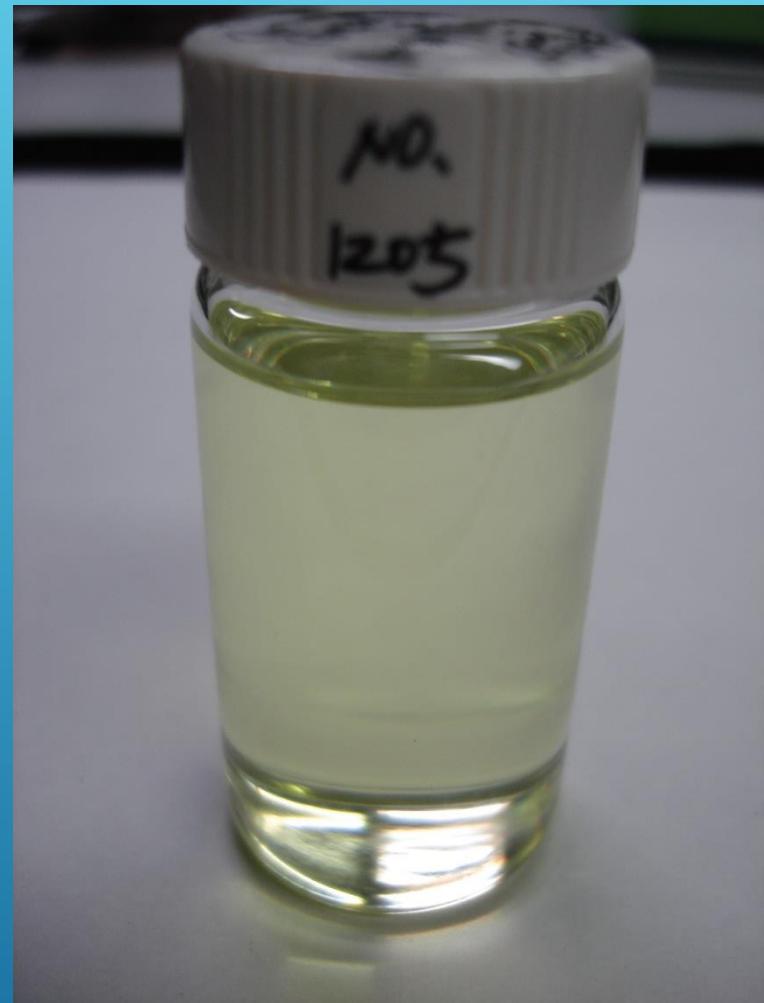
Yellow crystal



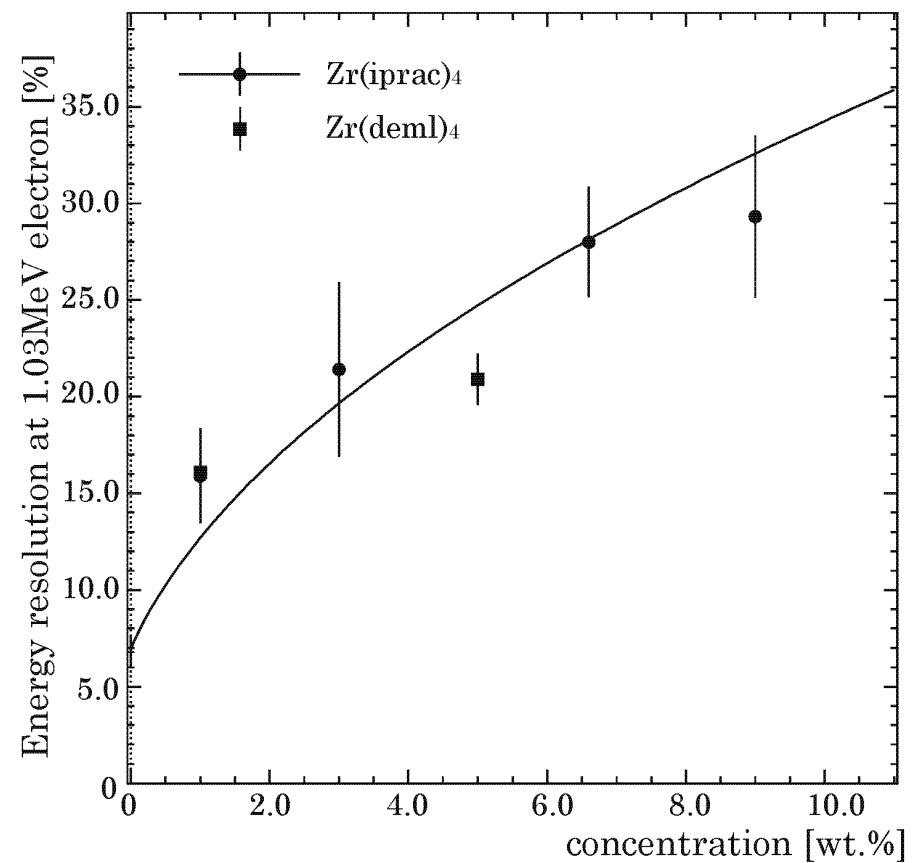
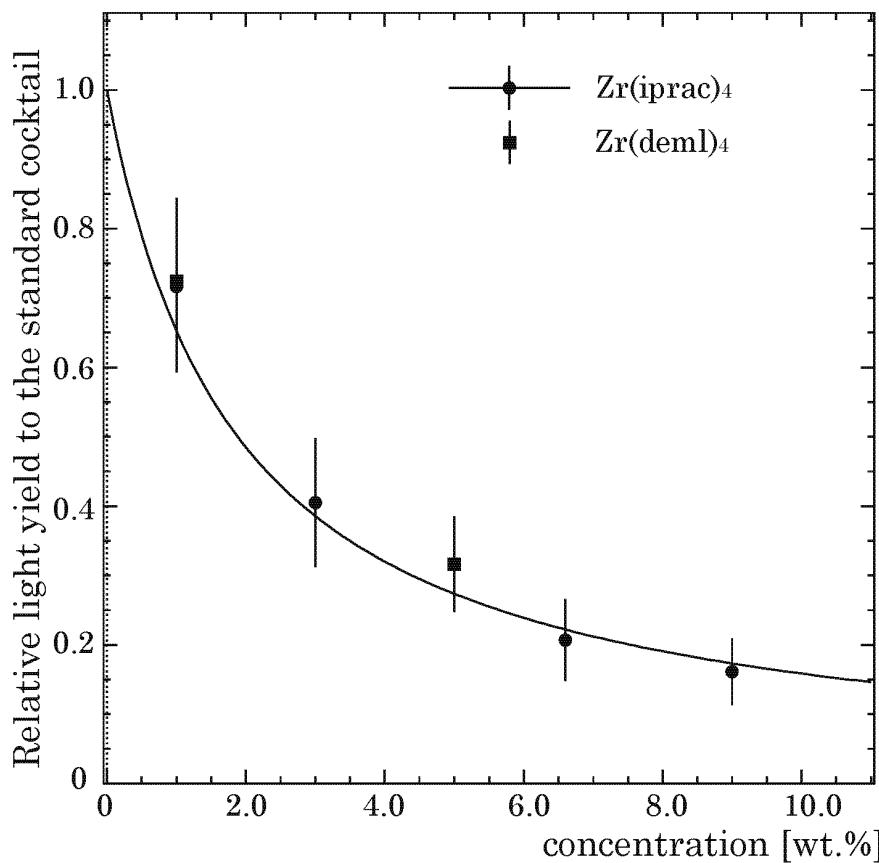
# Absorbance of Tetrakis (diethyl malonato) Zirconium



peak wavelength : 265.2nm



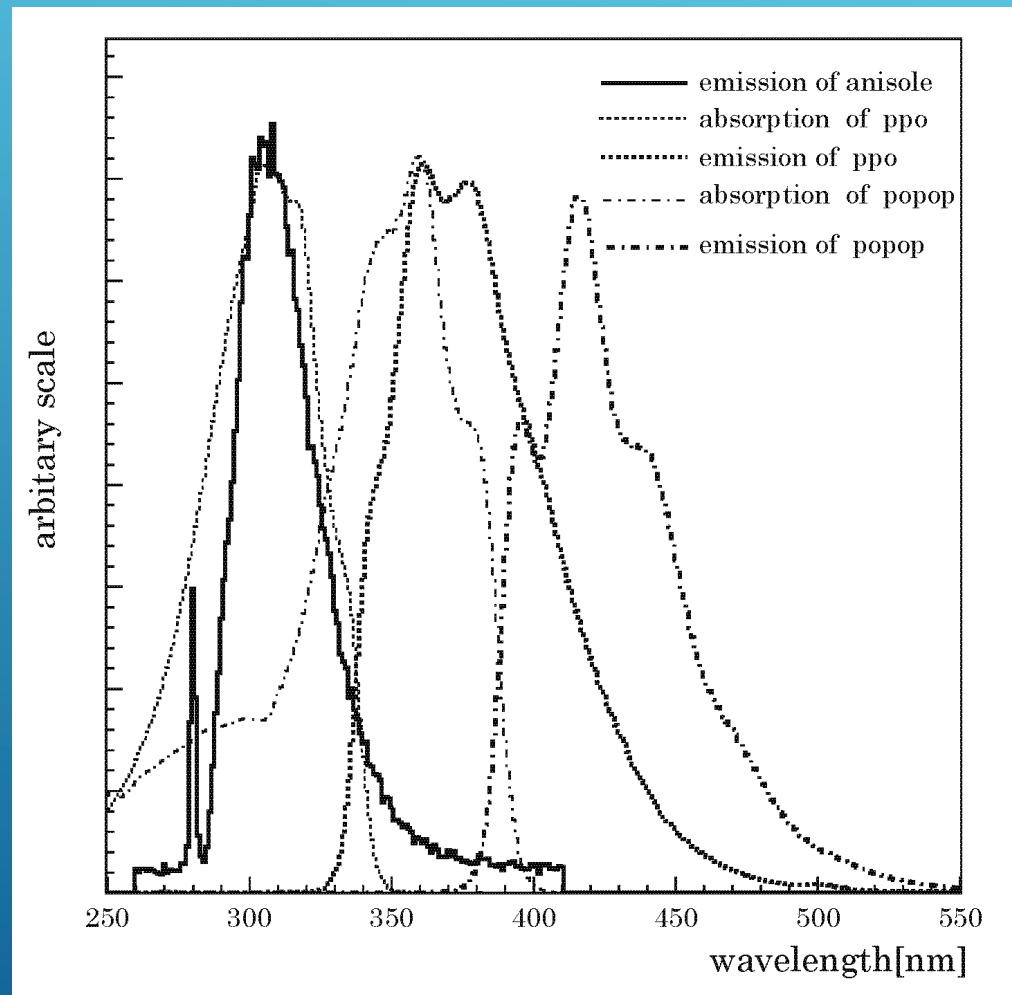
# Light yield and energy resolution of $\text{Zr}(\text{deml})_4$



$\text{Zr}(\text{deml})_4$  has a little better performance than  $\text{Zr}(\text{iprac})_4$ .

# BACKUP

# Emission and absorption spectra for solvent and solute in standard cocktail

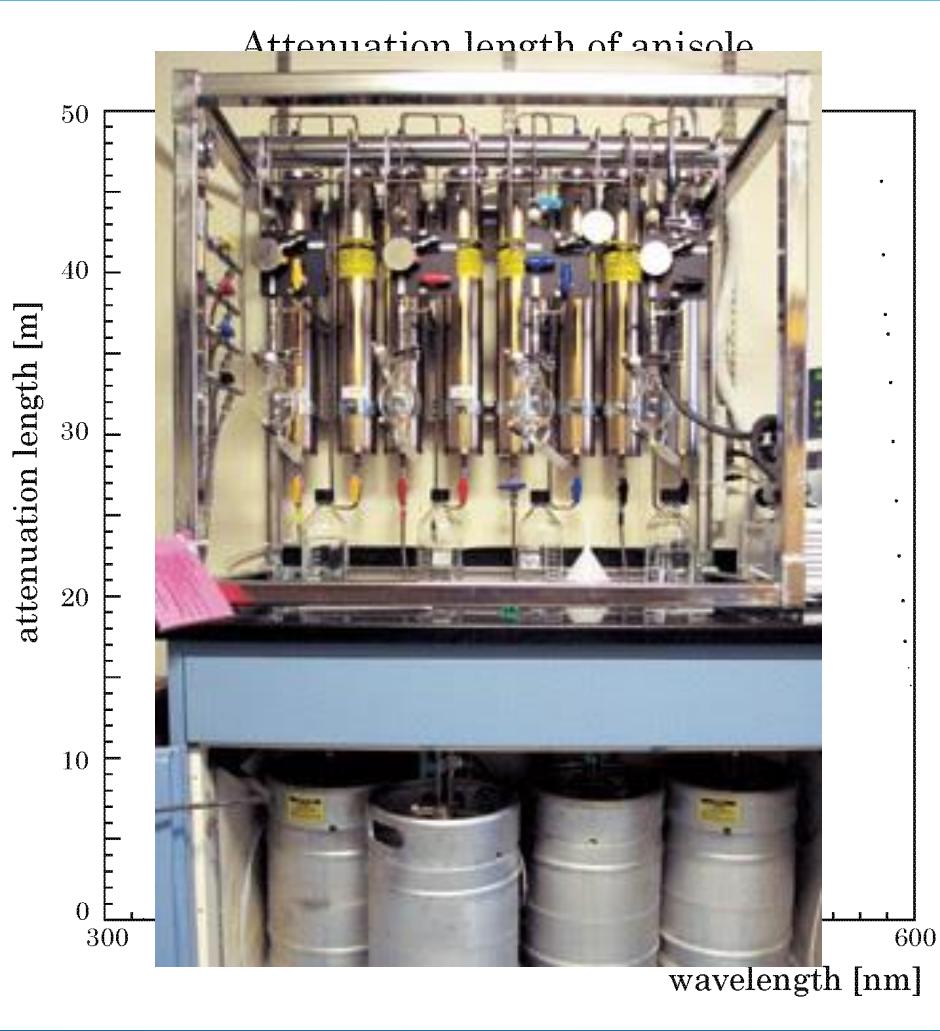


PPO absorbed most of emission lights from anisole.



Effectively the energy was transferred to the secondary scintillator.

# ATTENUATION LENGTH OF ANISOLE

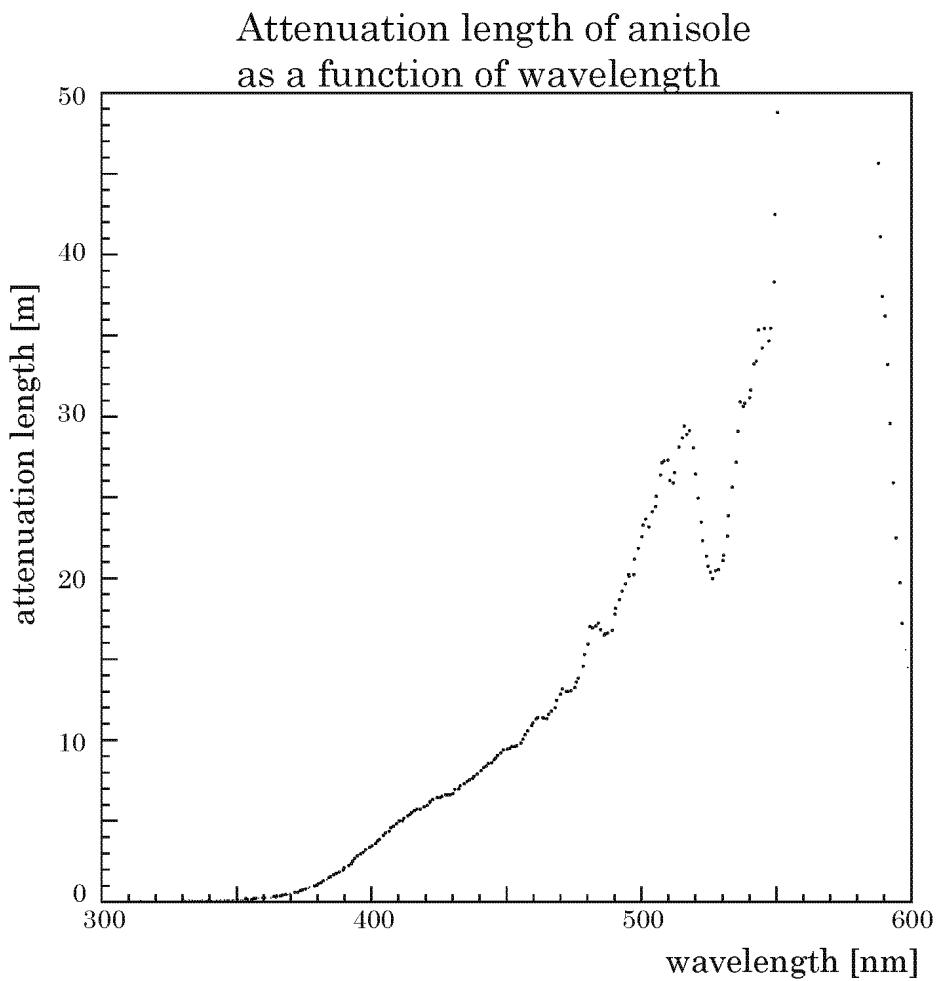


Attenuation length of light from POPOP was obtained as ~6m for current liquid scintillator.



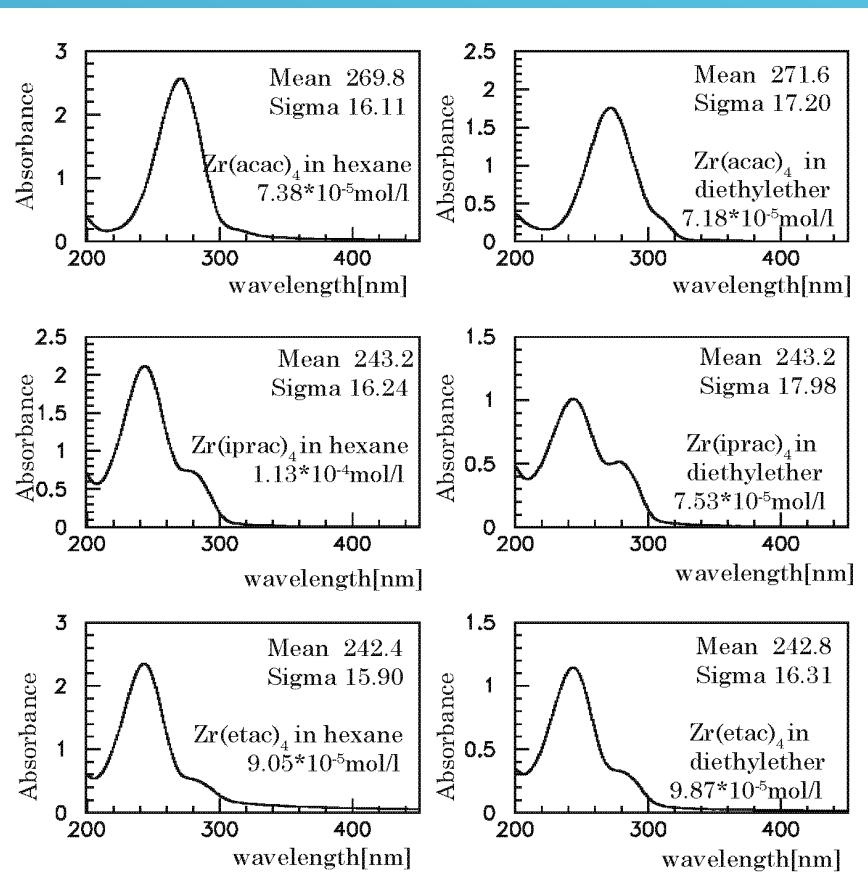
Attenuation length will be recovered ~15m by same purification method as RENO with  $\text{Al}_2\text{O}_3$ . (Ref: H.Grubbs et al., Org.Mat. 1996 15, 1518-1520)

# ATTENUATION LENGTH OF ANISOLE



Attenuation length of light from POPOP was obtained as ~6m. It is almost equivalent with the detector size.

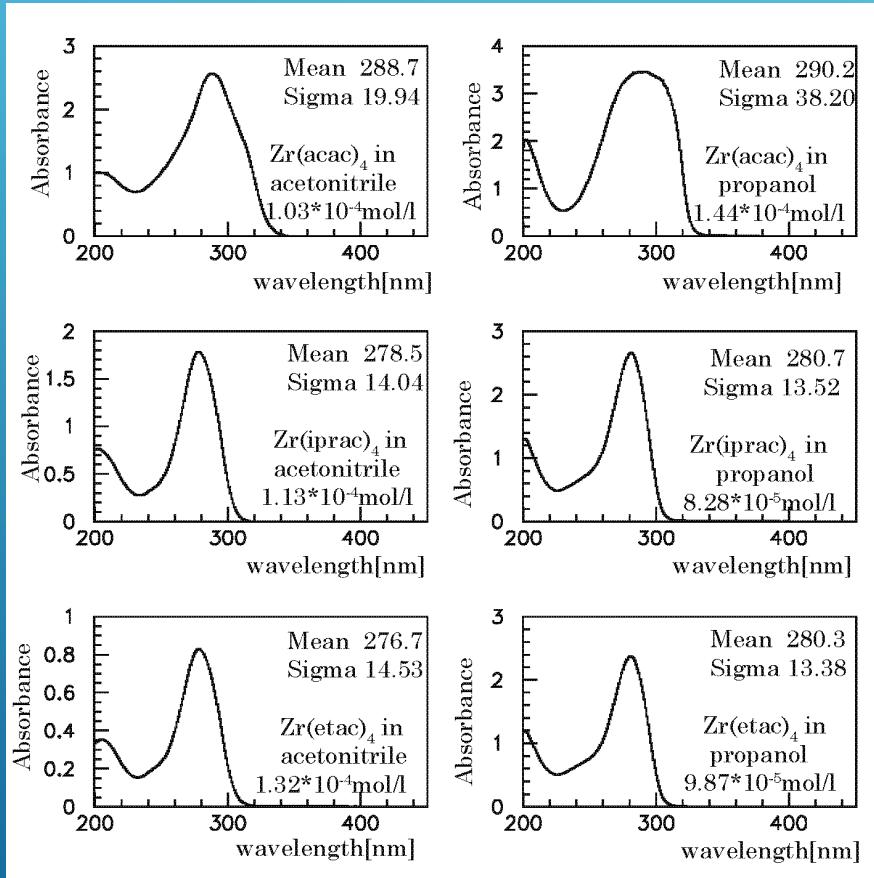
# Absorbance spectra in hexane / diethyl ether



~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.

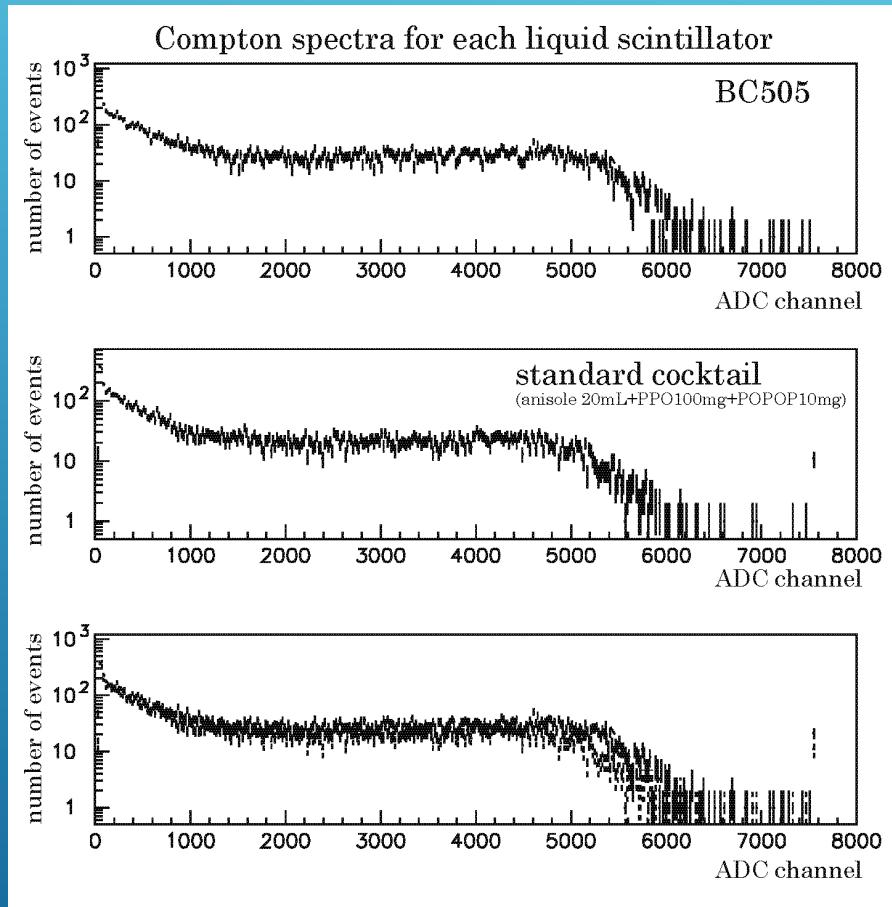


~280nm peak should be due to Zr(iprac)<sub>4</sub> and Zr(etac)<sub>4</sub>.

# Results from ICP Atomic Emission Spectrometry

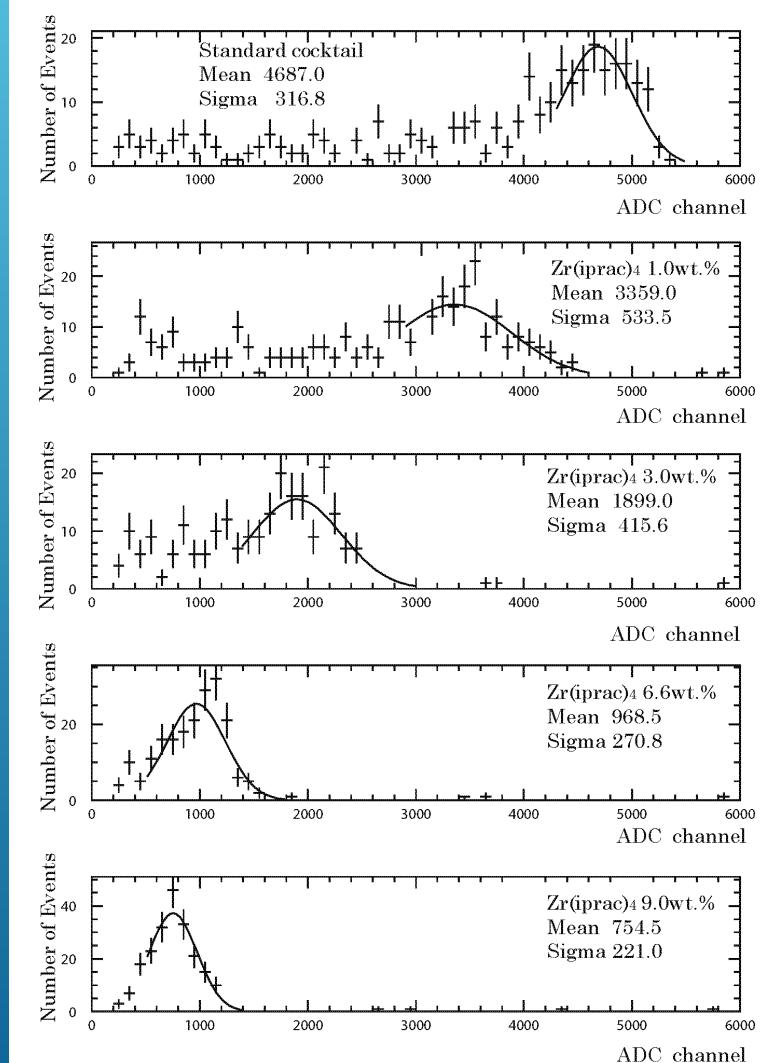
搬入日	試料名		ジルコニウム濃度 (mg/L)
平成26年11月28日	①	ジエチルエーテル Zr(iprac) <sub>4</sub> 8.3mg/20mL 上澄み液 2014.11.11 No.1075	0.50
	②	2014.10.17 Zr(iprac) <sub>4</sub> 5.9mg ジエチルエーテル No.980	150
平成26年12月10日	③	プロパノール Zr(iprac) <sub>4</sub> (1.5/20) 上5mL	5.6
	④	2-プロパノール(20mL)+Zr(iprac) <sub>4</sub> (1.5mg) 2013.11.22 No.497	18
	⑤	2-プロパノール+Zr(iprac) <sub>4</sub> 吸収1回目測定 2013.11.22 No.497から No.525	1.1
	⑥	アニソール Zr(iprac) <sub>4</sub> 2.0/20mL No.673(上)5mL	2.6

# 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.

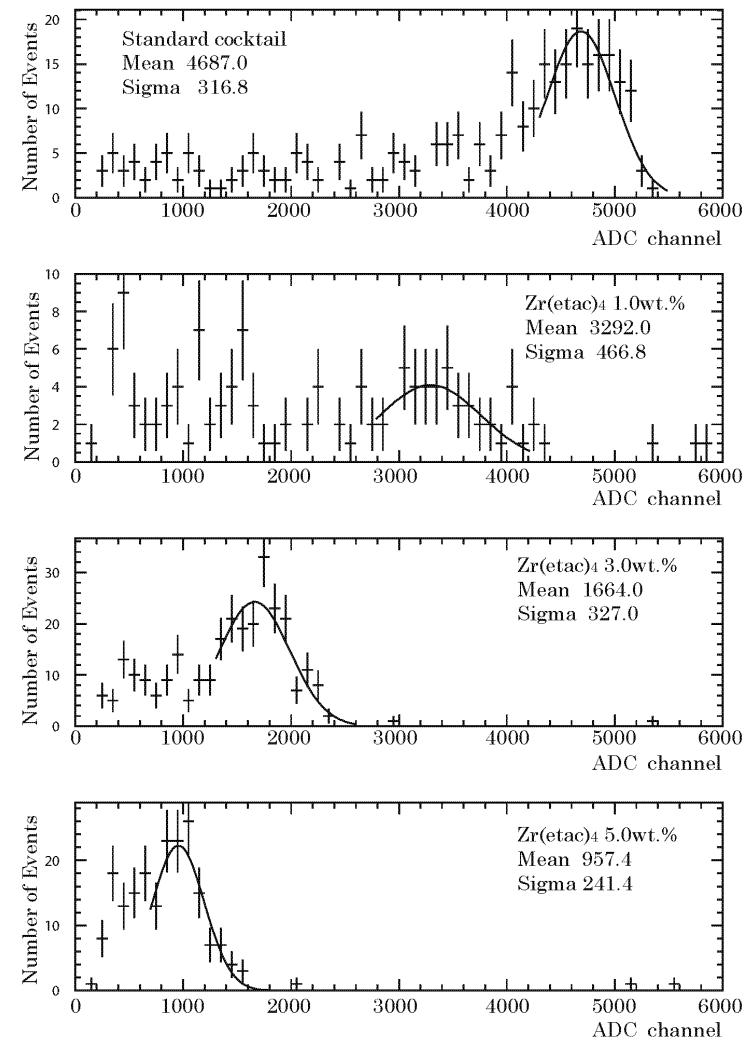
# ENERGY SPECTRA FOR SEVERAL CONCENTRATION OF ZR(IPRAC)4



Peak values decreased as a function of the concentration of Zr(iprac)<sub>4</sub>.

Energy resolutions are also getting worse as a function of the concentration of Zr(iprac)<sub>4</sub>.

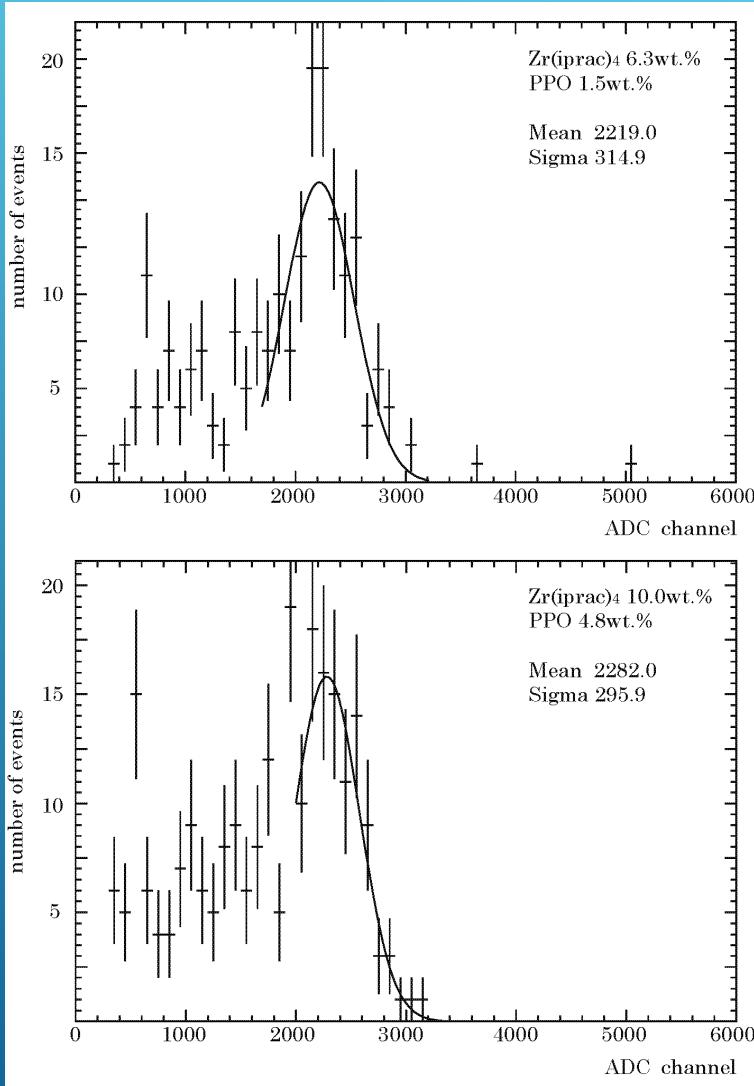
# ENERGY SPECTRA FOR SEVERAL CONCENTRATION OF ZR(ETAC)4



Peak values decreased as a function of the concentration of Zr(etac)<sub>4</sub>.

Energy resolutions are also getting worth as a function of the concentration of Zr(etac)<sub>4</sub>.

# RECOVERY FOR ABILITY OF LIGHT YIELD AND ENERGY RESOLUTION

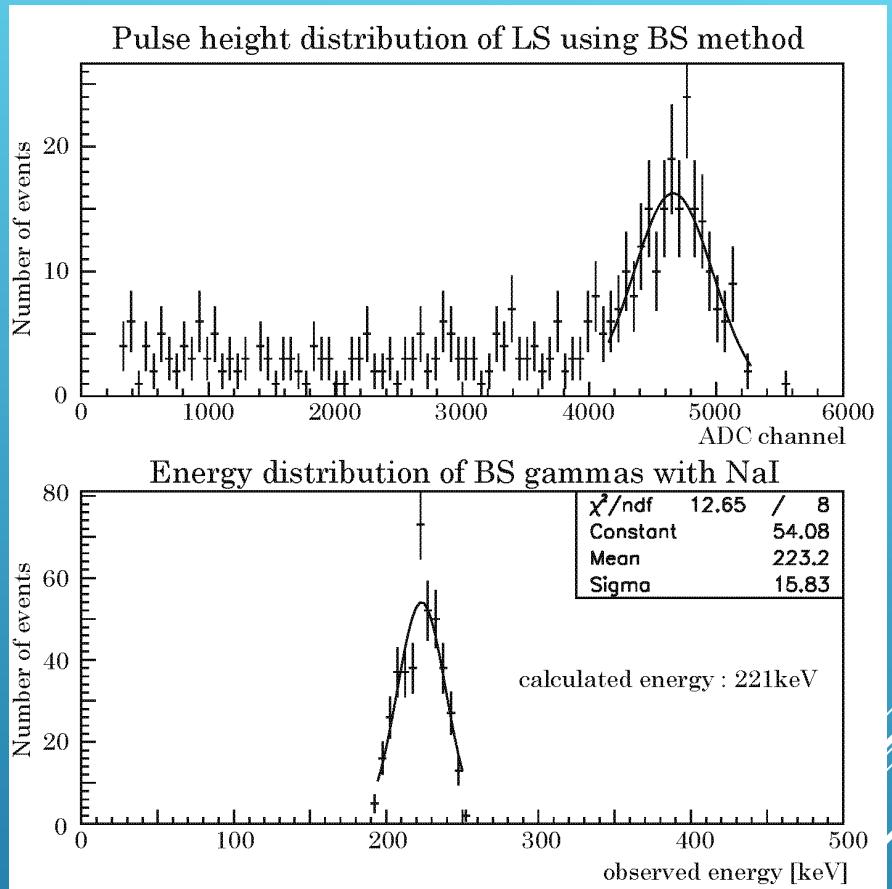
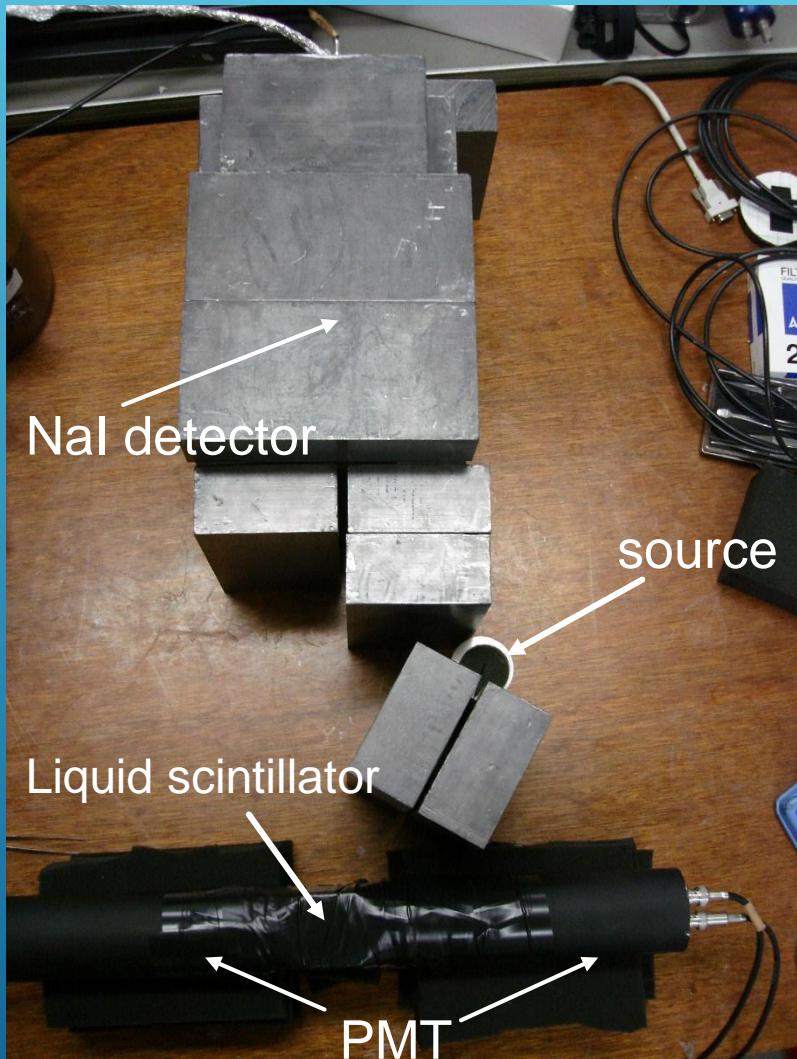


PPO helps recovering the light yield and the energy resolution.



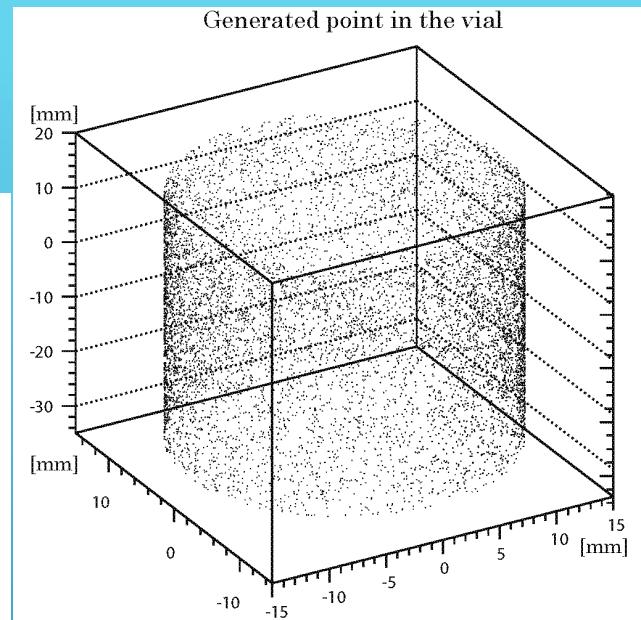
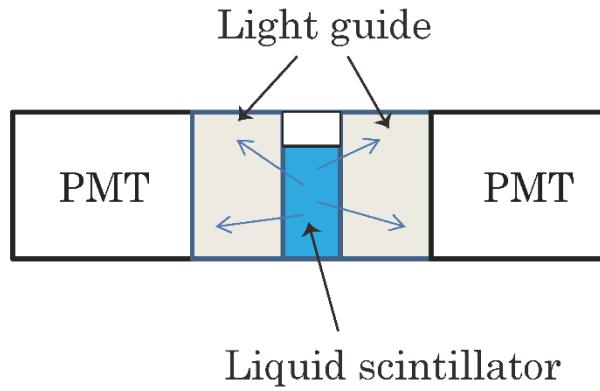
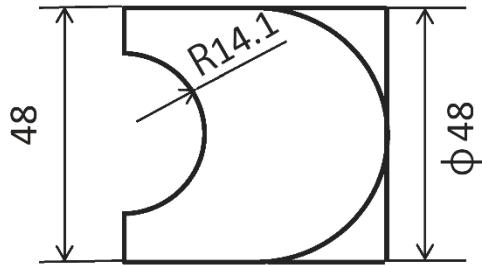
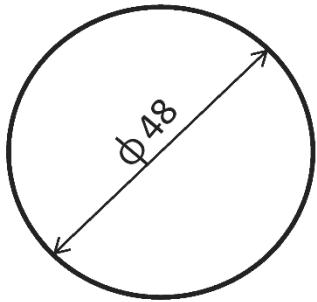
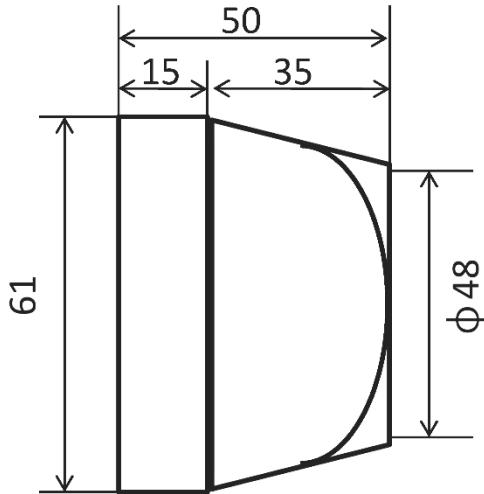
Confirmed our assumption and obtained optimized real cocktail (PPO 5wt.% POPOP 0.5wt.%)

# BACK SCATTERING METHOD

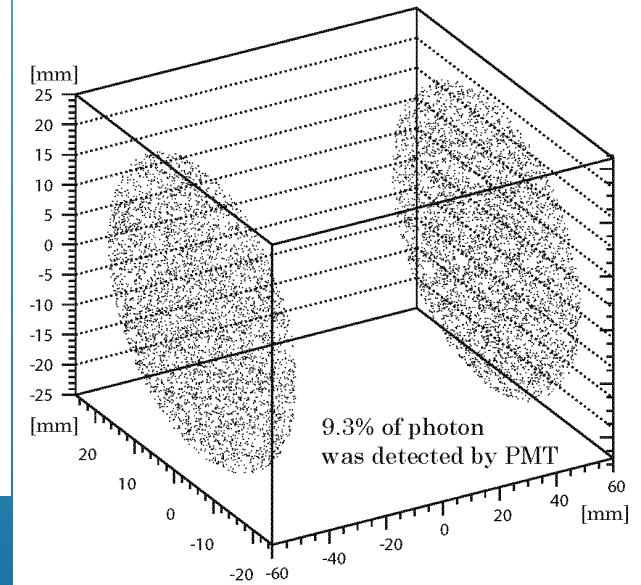


Single peak could be used even in liquid scintillator.

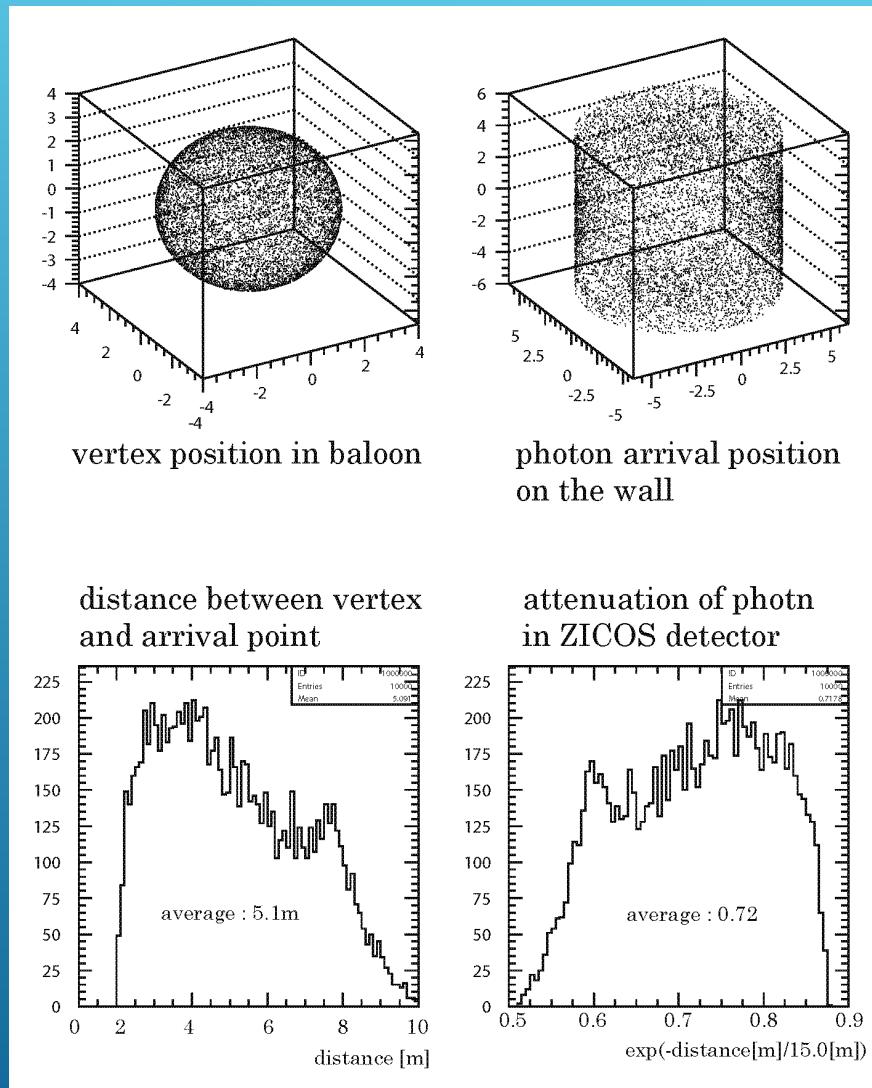
# Photo coverage



detected scintillation light on PMT surface

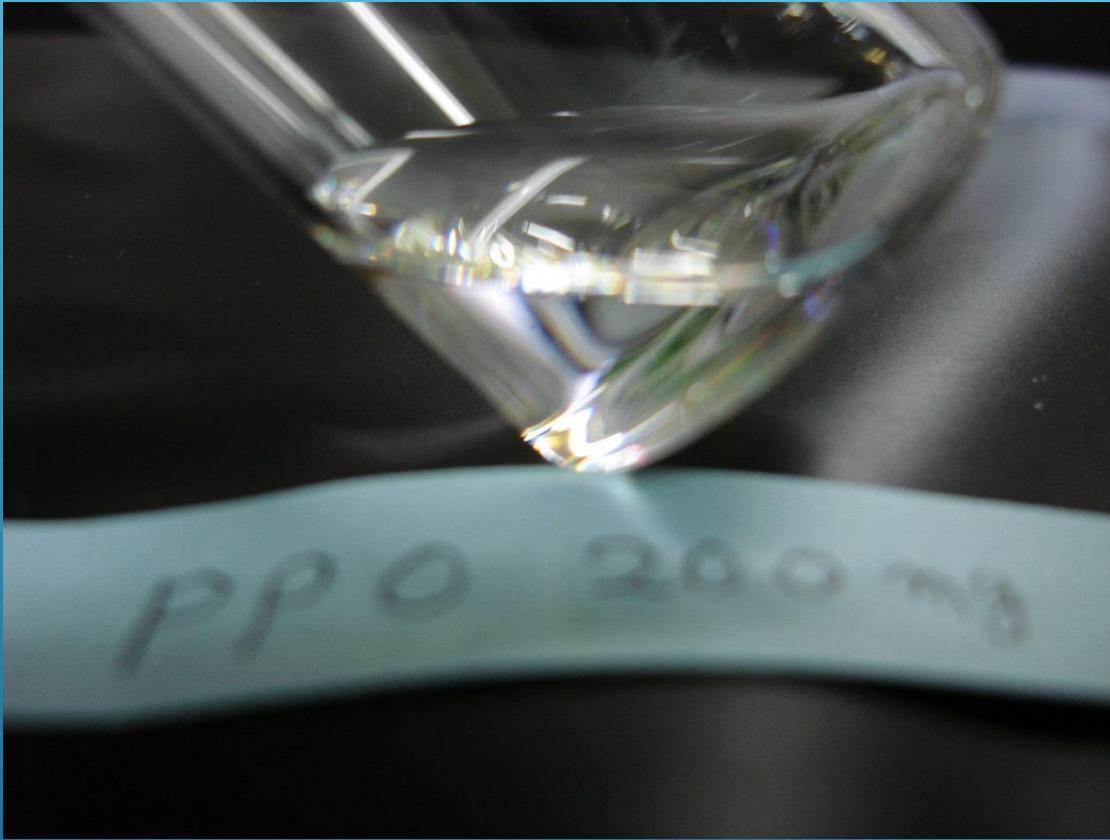


# Photon attenuation in ZICOS detector



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)<sub>4</sub>



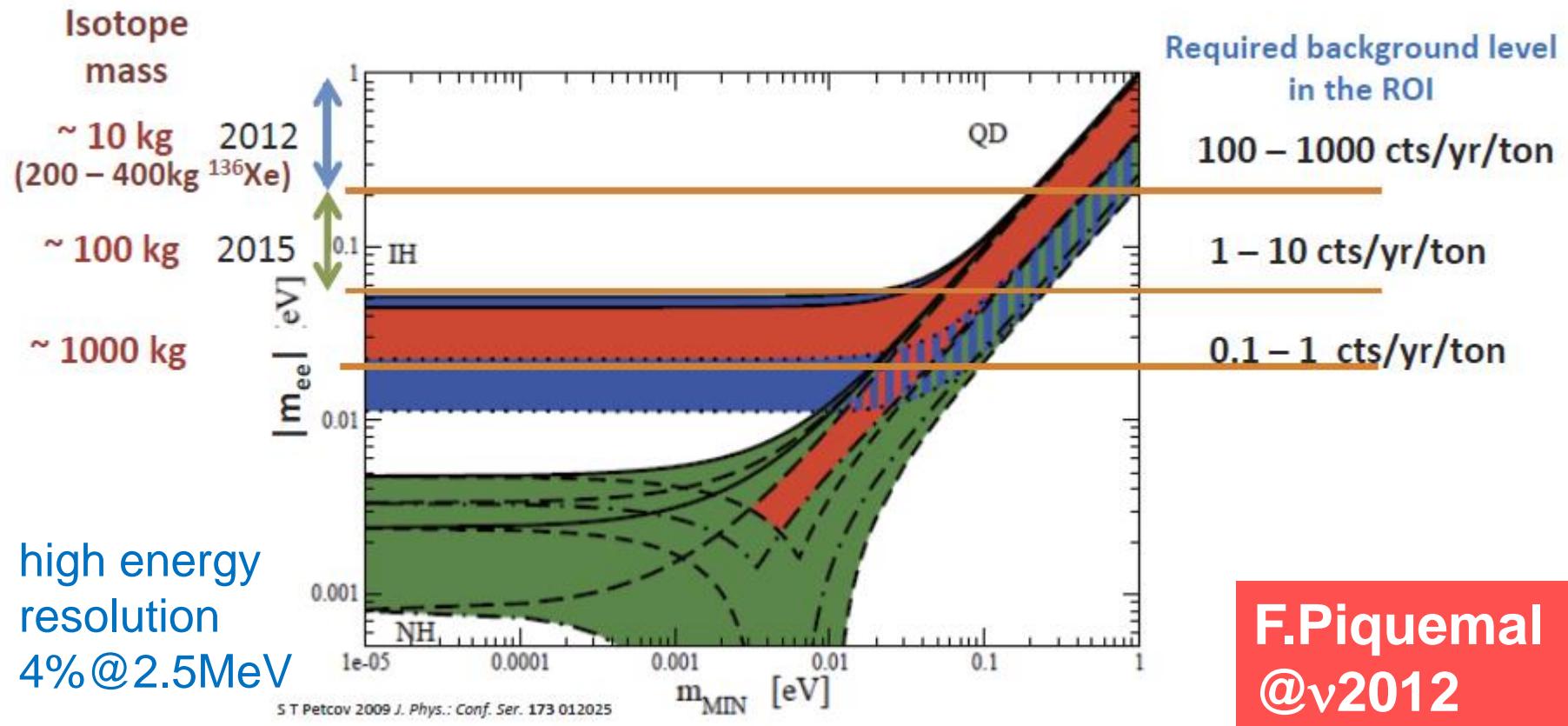
$$\frac{205.5\text{mg}}{205.5\text{mg}+2.0\text{g}}$$

$$= 9.3 \text{ wt.\%}$$



Maybe solved in  
20 wt.\% of Zr(iprac)<sub>4</sub>

# FOR FUTURE EXPERIMENTS



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

~tons of target will be necessary for next generation detector