

ZICOS – NEW PROJECT FOR NEUTRINOLESS DOUBLE BETA DECAY EXPERIMENT USING ZIRCONIUM COMPLEX IN ORGANIC LIQUID SCINTILLATOR –

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Revealing the history of the universe with
underground particle and nuclear research 2016

Koshiha Hall, Tokyo

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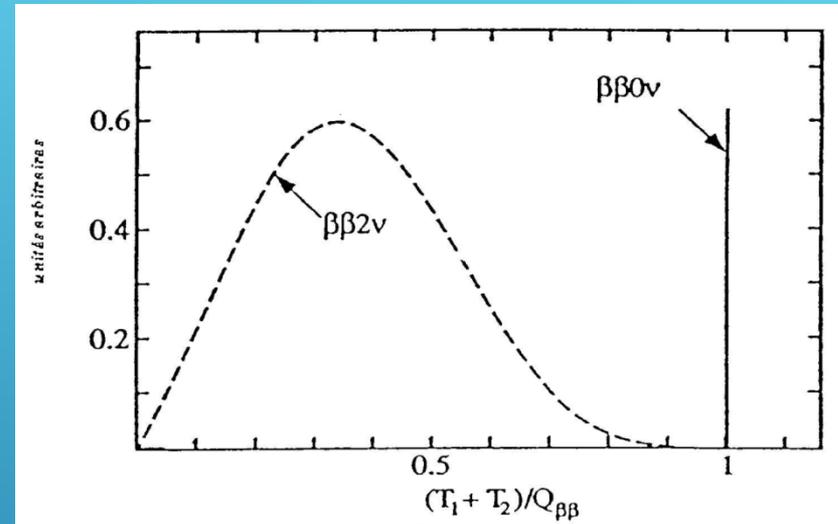
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Neutrinoless double beta decay

$\beta\beta$ emitters with $Q_{\beta\beta} > 2$ Mev

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



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

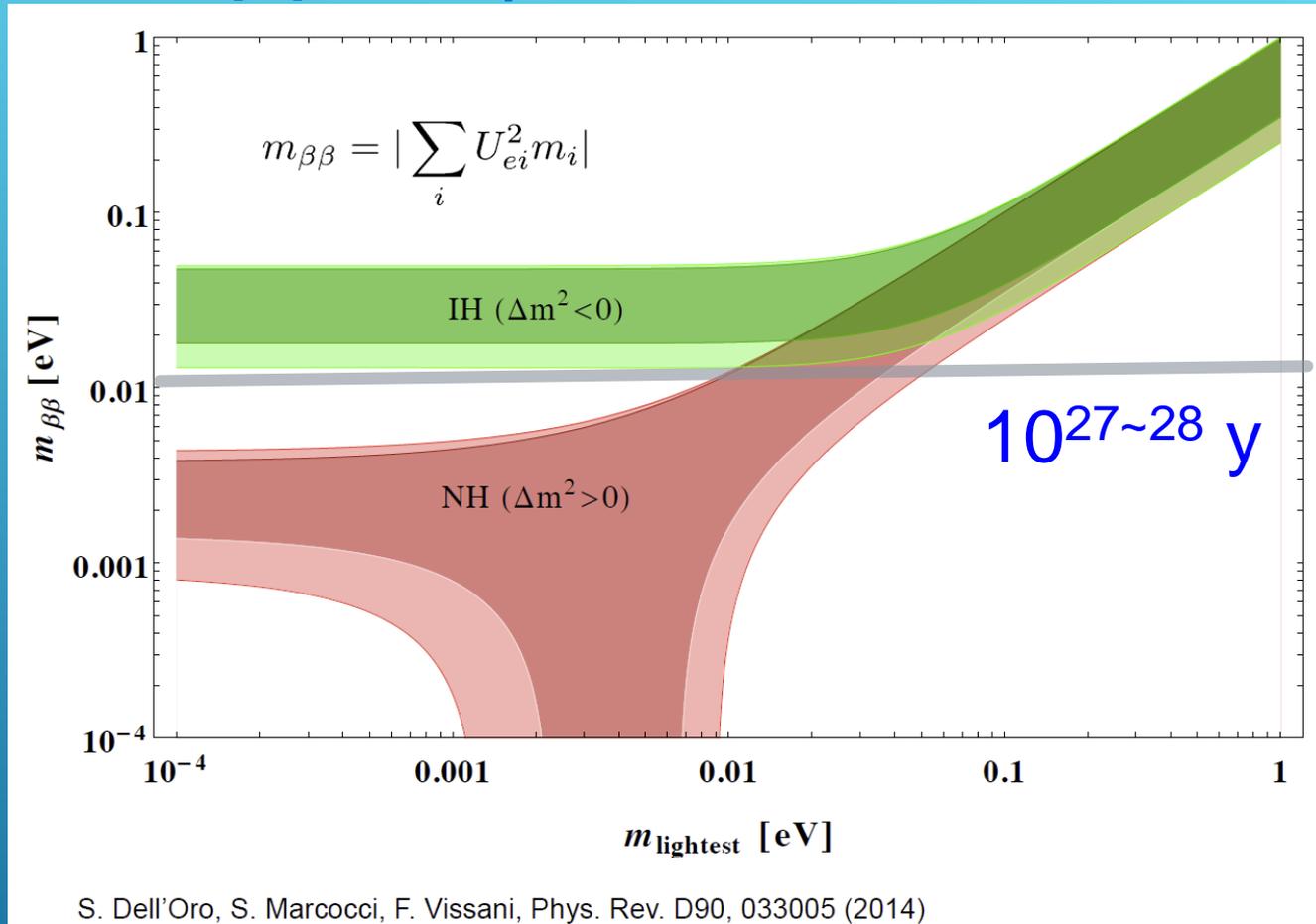
$$T_{1/2} \sim a(Mt/\Delta E \cdot B)^{1/2}$$

a: abundance M: target mass

t: measuring time ΔE : energy resolution B: BG rate

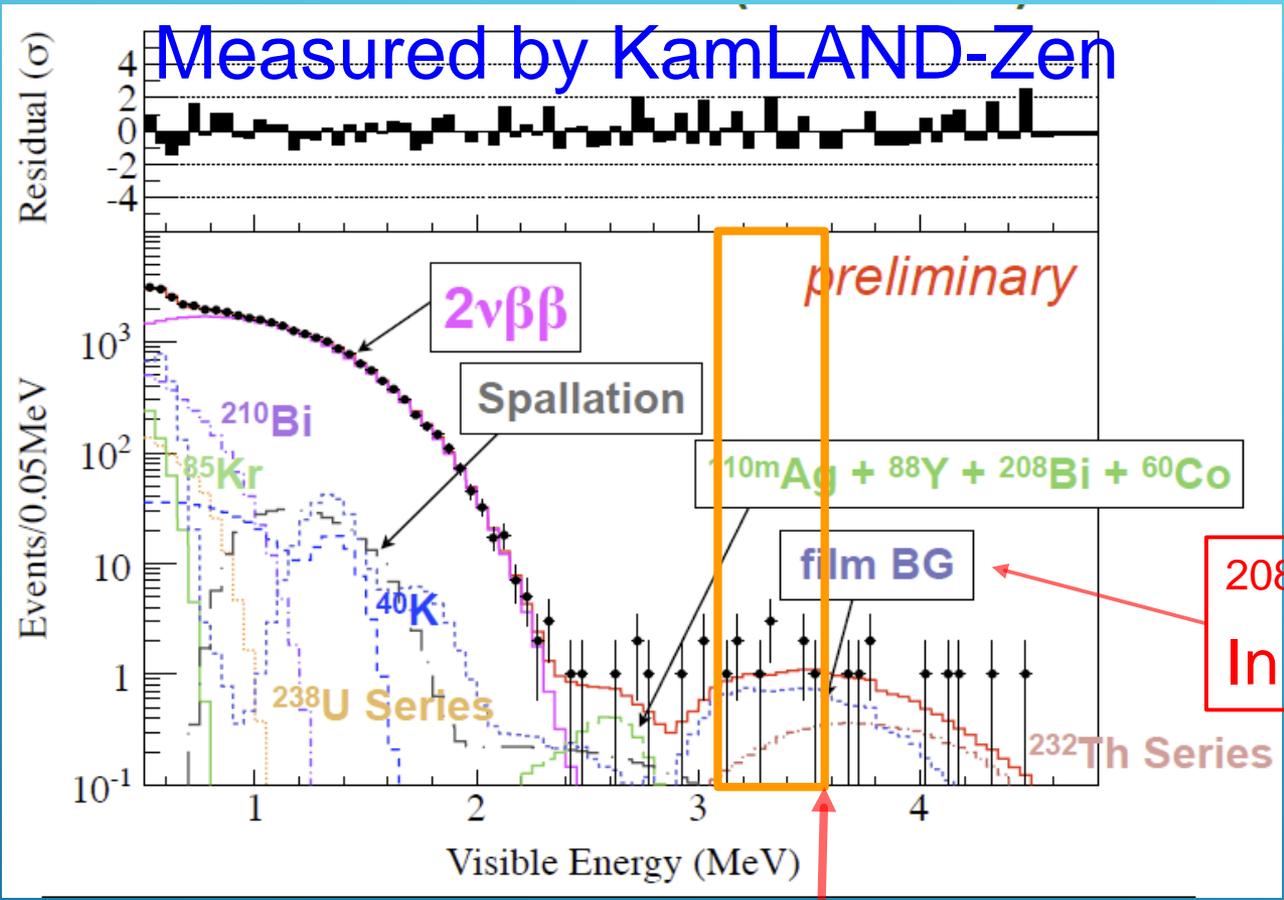
Requirement : Low BG, Large target mass, High energy resolution

Future $0\nu\beta\beta$ experiments



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

Backgrounds around 3.35MeV



I.Shimizu@Neutrino2014

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

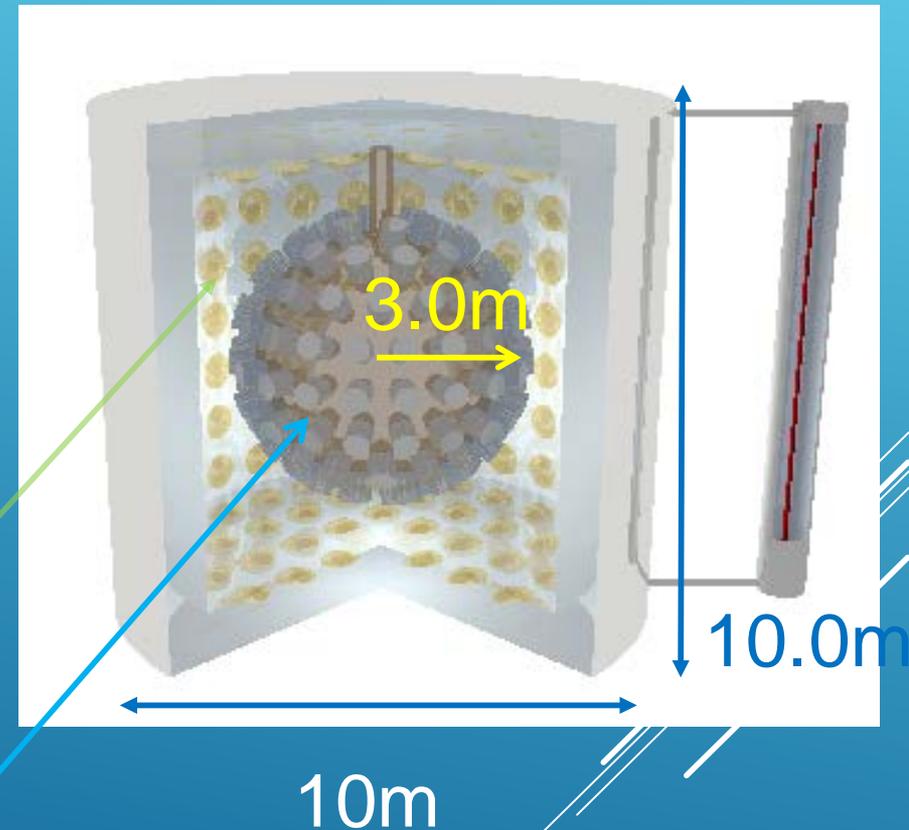
ZICOS- Zirconium Complex in Organic Liquid Scintillator for neutrinoless double beta decay

Goals for development of LS :

- (1) Zr should be solved highly in order to make detector small.**
- (2) 3.5% at 3.35MeV of energy resolution, if ZICOS has PMTs with 40% photo coverage and long attenuation length (~10m)**

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

Inner detector with 40% photo coverage 10" PMT including Zirconium loaded 113 tons LS



Neutrino mass sensitivity of ZICOS experiment

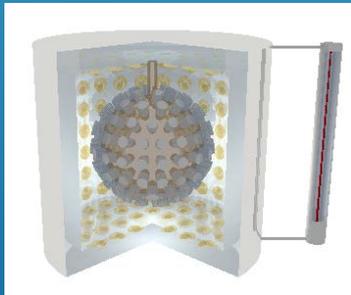
Results from NEMO-3 (^{96}Zr) : $T_{1/2}^{0\nu} > 9.2 \times 10^{21}\text{y}$
 $\langle m_{\nu} \rangle$ 7.2 – 10.8 eV ($g_A=1.25, g_{pp}=1.11, \text{QRPA}$)

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

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

($T_{1/2}^{0\nu} > 2.6 \times 10^{25}\text{y}$) (Ref: I.Shimizu arXiv:1409.0077 (2014))

Mass : 113 ton



10wt.% $\text{Zr}(\text{iprac})_4 = 12.6\text{ton}$
includes 1.7ton of Zirconium

= 45 kg of ^{96}Zr (natural abundance 2.6%)
(64kg of $^{136}\text{Xe} = 0.2 \times \text{KL-Zen}$)

$T_{1/2}^{0\nu} > 1.2 \times 10^{25}\text{y}$ ← 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 ton (4.4 times ^{136}Xe 320kg)

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

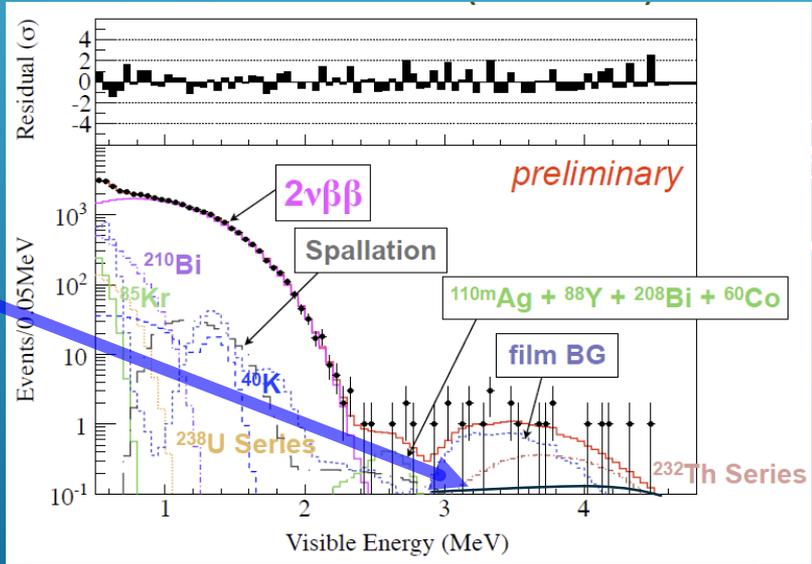
Electromagnetic isotope separation is expensive...

2) Lowering BG (^{208}Tl / ^{214}Bi)

i.e. $< 1/20 \times \text{KL-Zen}$
($\sim 1.0 \text{ events/ton/year}$)

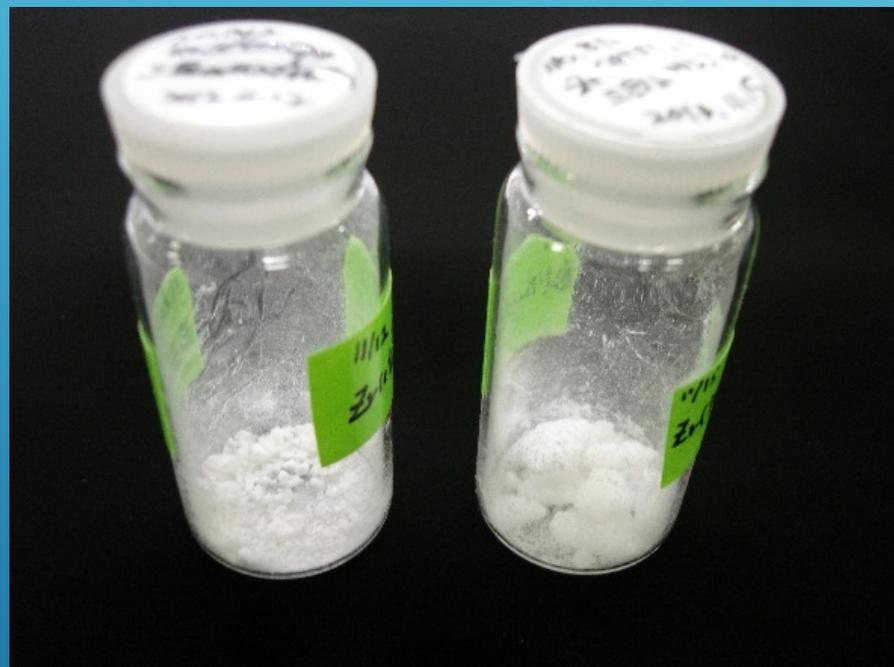
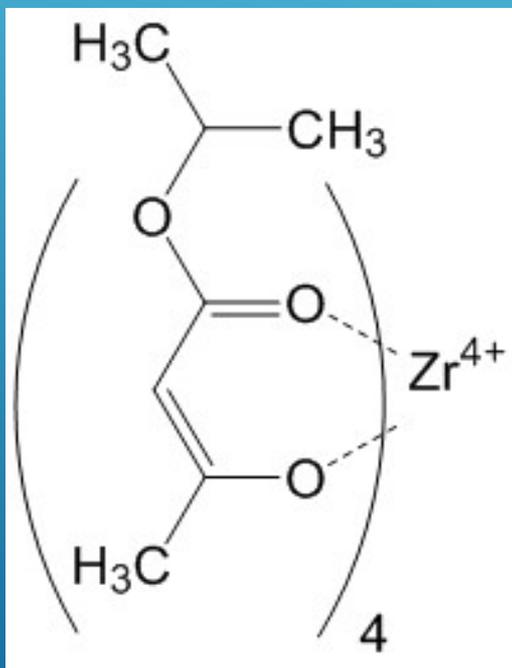
$T_{1/2}^{0\nu} > 5 \times 10^{25}\text{y}$

Maybe we can.....



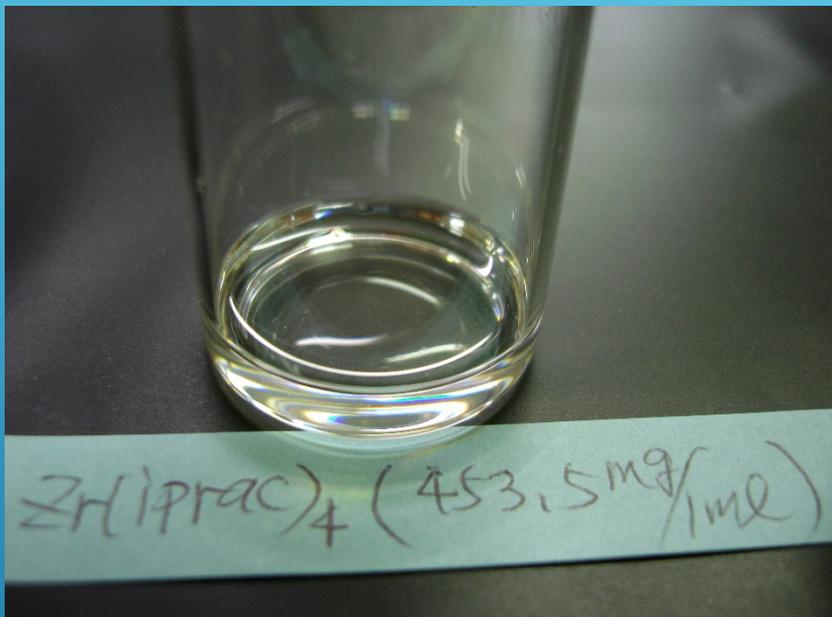
Development of Zr loaded LS

$\text{Zr}(\text{CH}_3\text{COCHCOOCH}(\text{CH}_3)_2)_4$: $\text{Zr}(\text{iprac})_4$
tetrakis (isopropyl acetoacetate) zirconium
mw : 663.87



Solid crystal or powder

Solubility of $Zr(iprac)_4$ for anisole

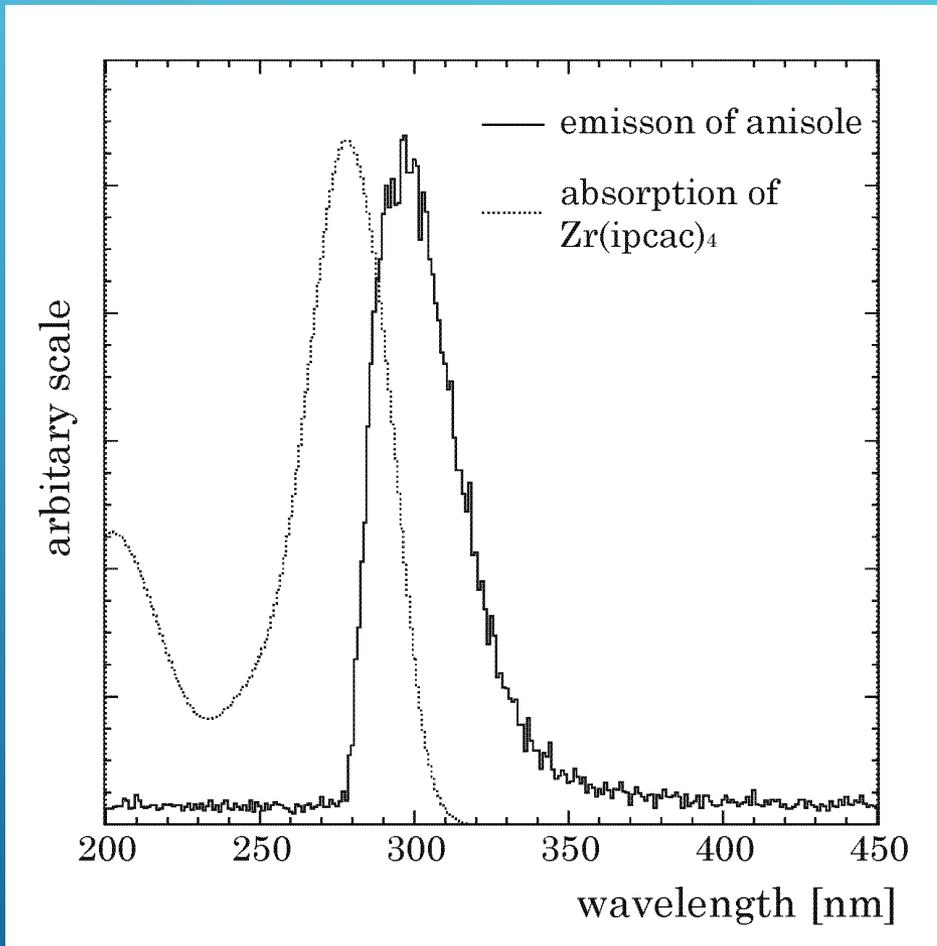


Solubility > 31.2 wt.%

$Zr(iprac)_4$ 2242mg, PPO 999mg
and POPOP 10mg solved in
20mL Anisole

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

Absorbance spectra for $Zr(iprac)_4$



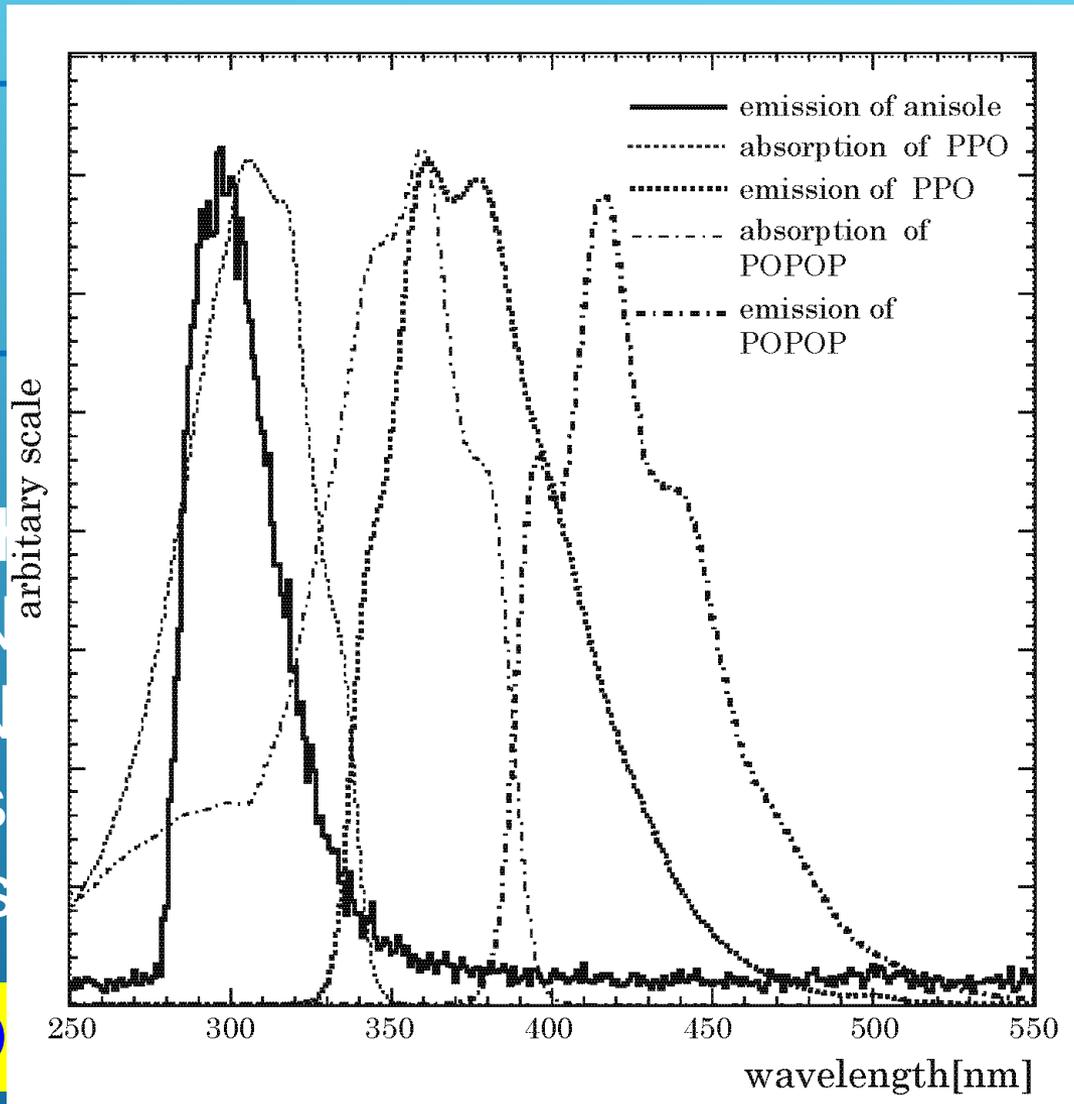
Absorption peaks of $Zr(iprac)_4$ was found around at 278nm. However, overlapped region with emission of anisole was existed.



$Zr(iprac)_4$ works as a quencher for the liquid scintillator system.

Light yield quenching by $Zr(iprac)_4$

L_0 : Light
 N_{ppo} : N
 N_{Zr} : N
 σ_1 : abs
 σ_2 : abs

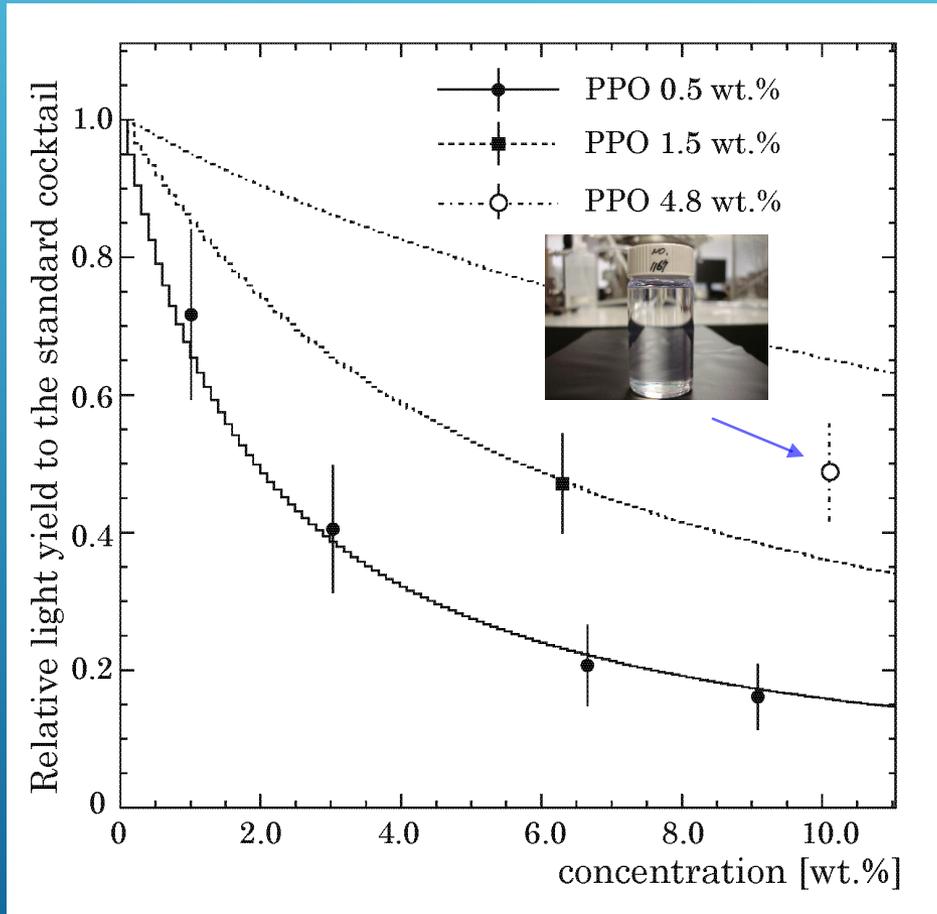


PPO

ield.

Recovering the light yield

Measured at several conditions of PPO concentration



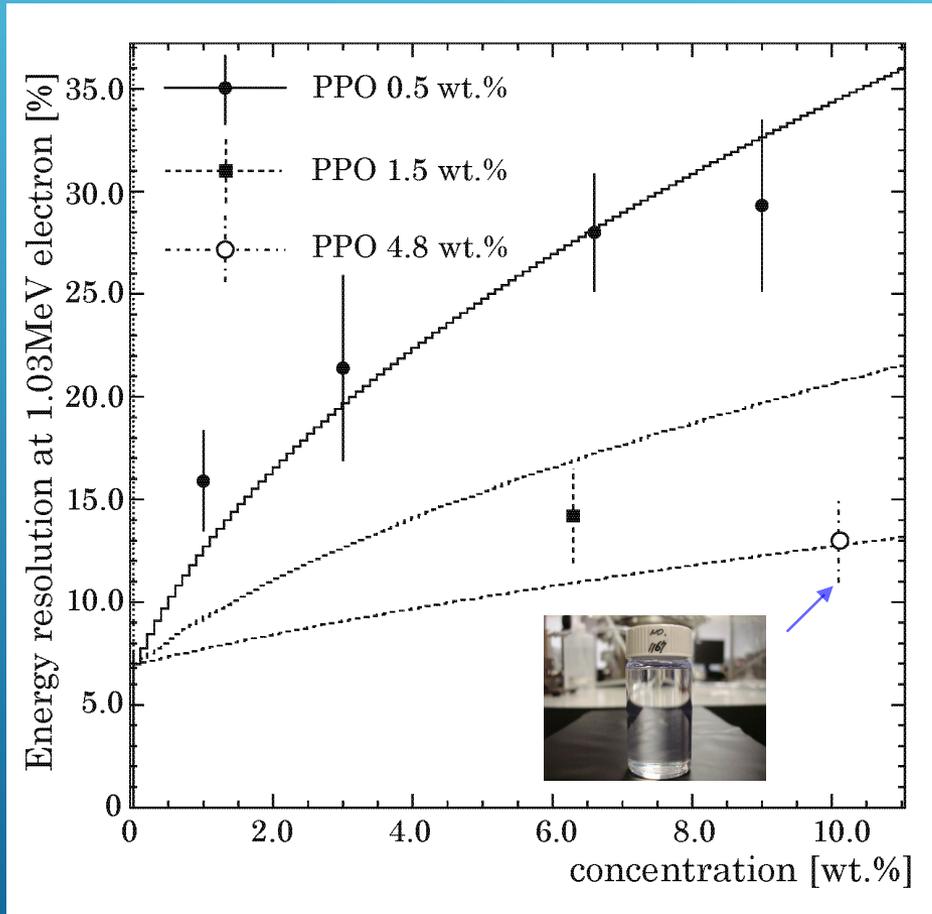
5wt.% PPO helps actually recovering the scintillation light yield.



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 $Zr(iprac)_4$.

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

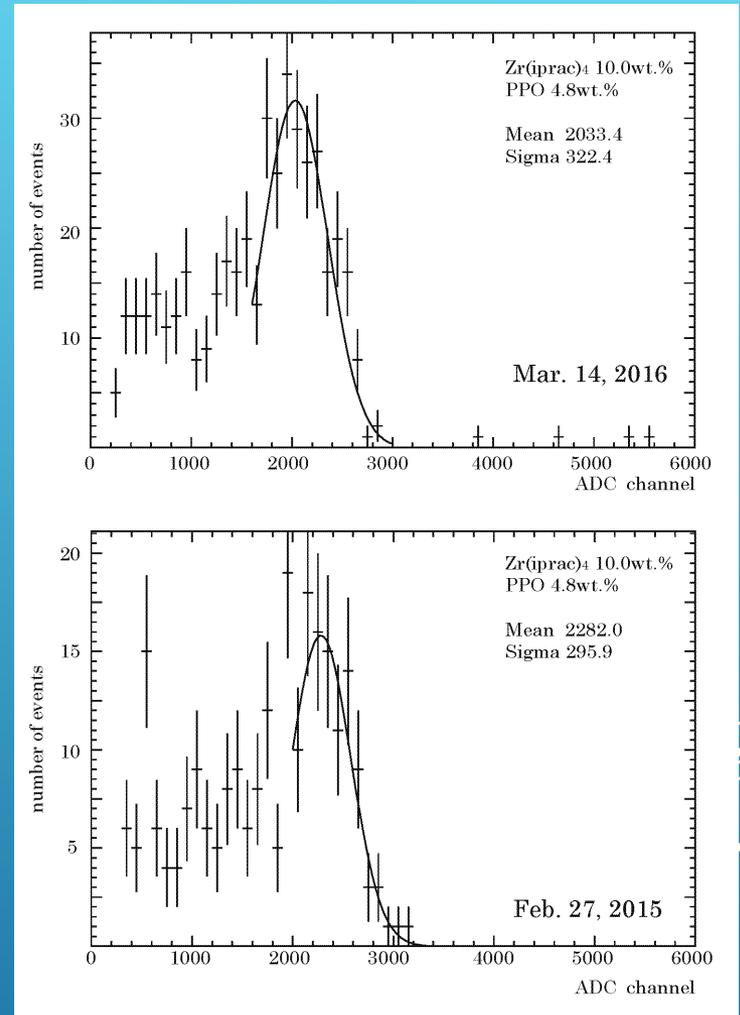
Achieved goal!

Stability of liquid scintillator

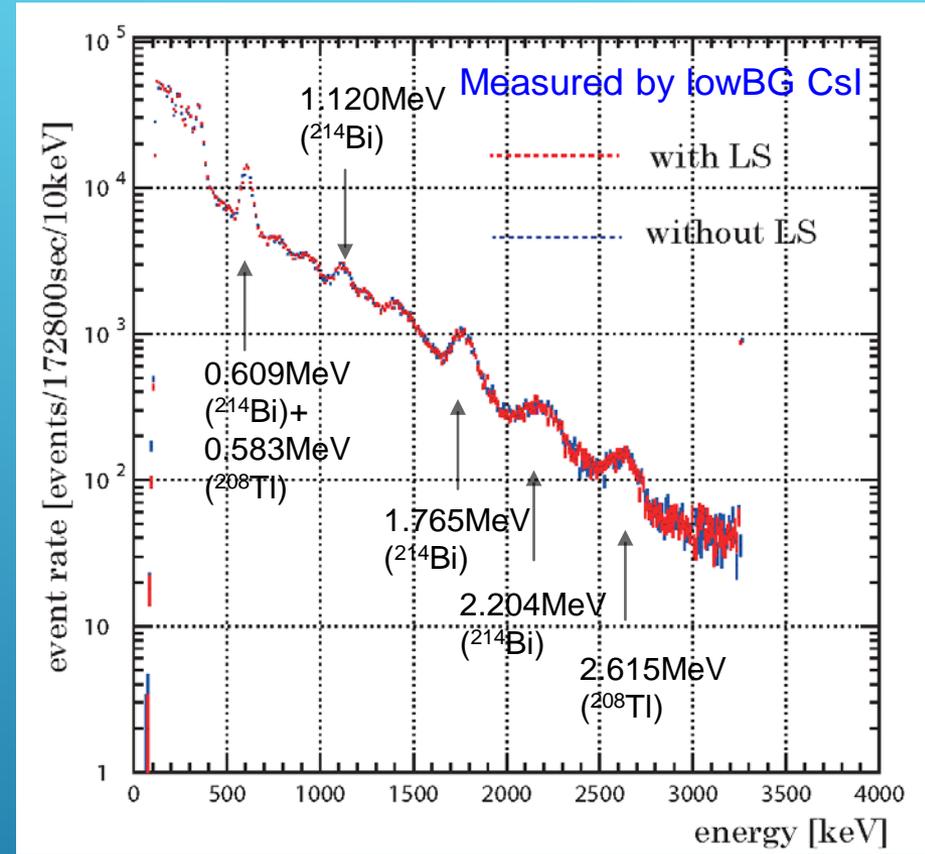


Feb. 27, 2015 Mar. 14, 2016

**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} g/g)

Main backgrounds for ZICOS

Main backgrounds evaluated by KamLAND-Zen are mainly ^{208}Tl inside the balloon film.

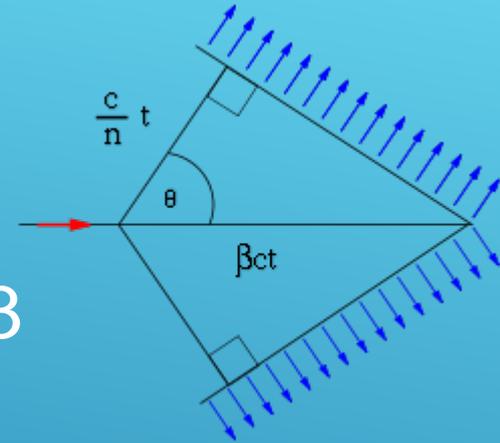


Need additional technique except energy spectral shape obtained by scintillation light in order to reduce those backgrounds drastically.

Can we use Cherenkov lights for background reduction ?

Property of Cherenkov light

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

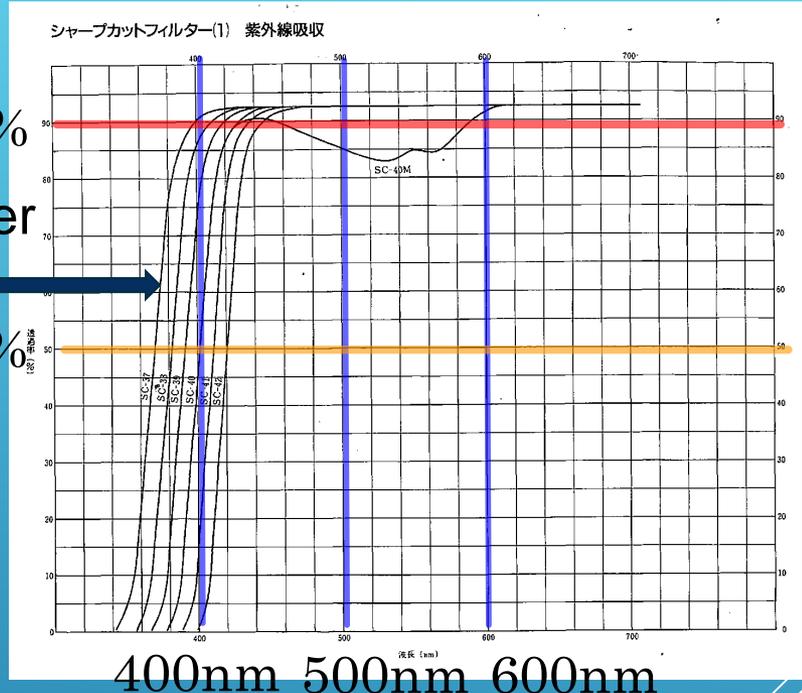
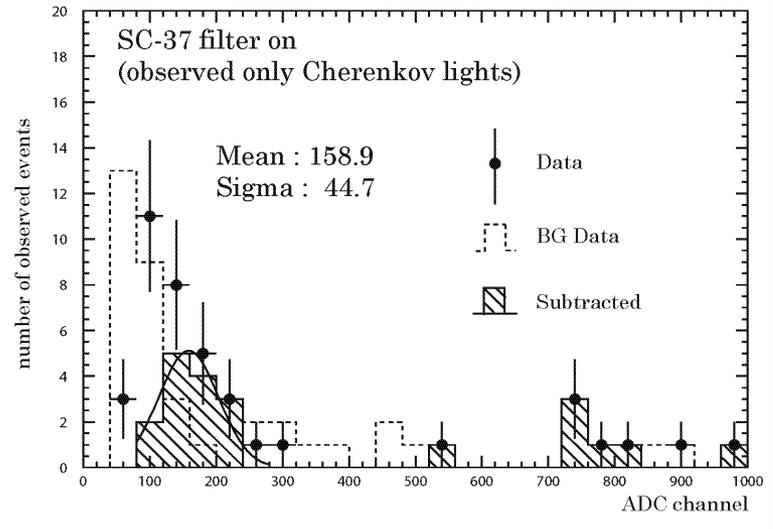
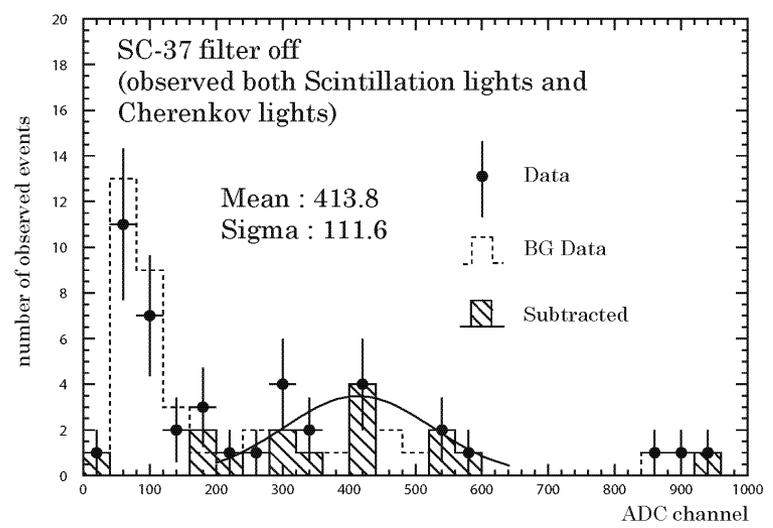


$$\frac{dN}{dx} = 2\pi z^2 \alpha \sin^2 \theta_c \int_{\lambda_1}^{\lambda_2} \frac{d\lambda}{\lambda} = 475 z^2 \sin^2 \theta_c \text{ photon/cm}$$

c.f. Light yield of Scintillation : ~ 12000 photon/MeV

Cherenkov light = $\sim 1\%$ of scintillation light

Measurement of LY for Cherenkov lights



Cherenkov light yield ($\lambda > 400\text{nm}$)
 Scintillation light yield of std. LS

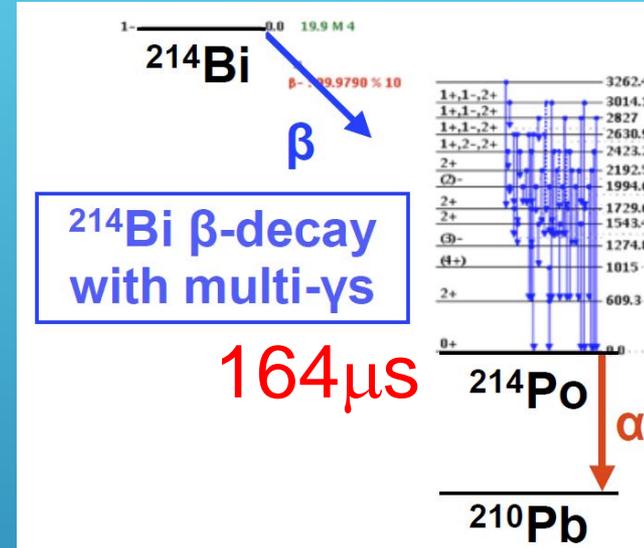
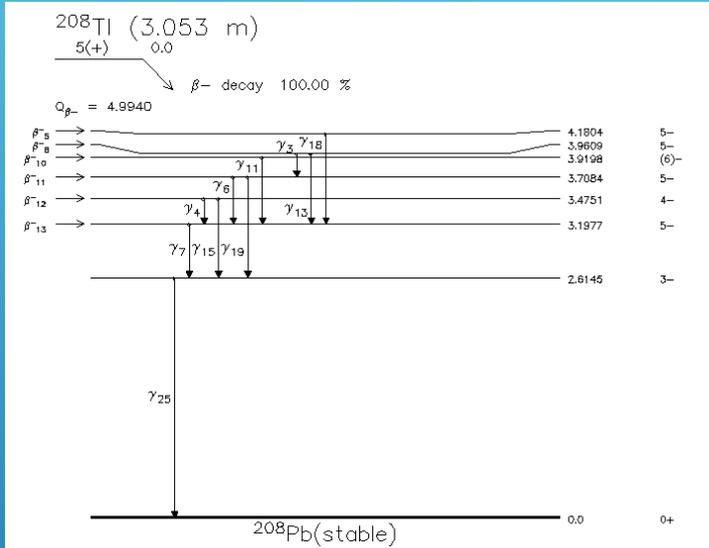
$$= \frac{159/0.15/0.95}{255/0.046/0.098} = \sim 0.02$$

Consistent with expectation.

Significant backgrounds

^{208}Tl : β and 2.6MeV γ s

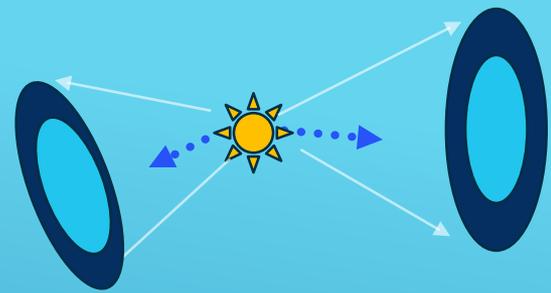
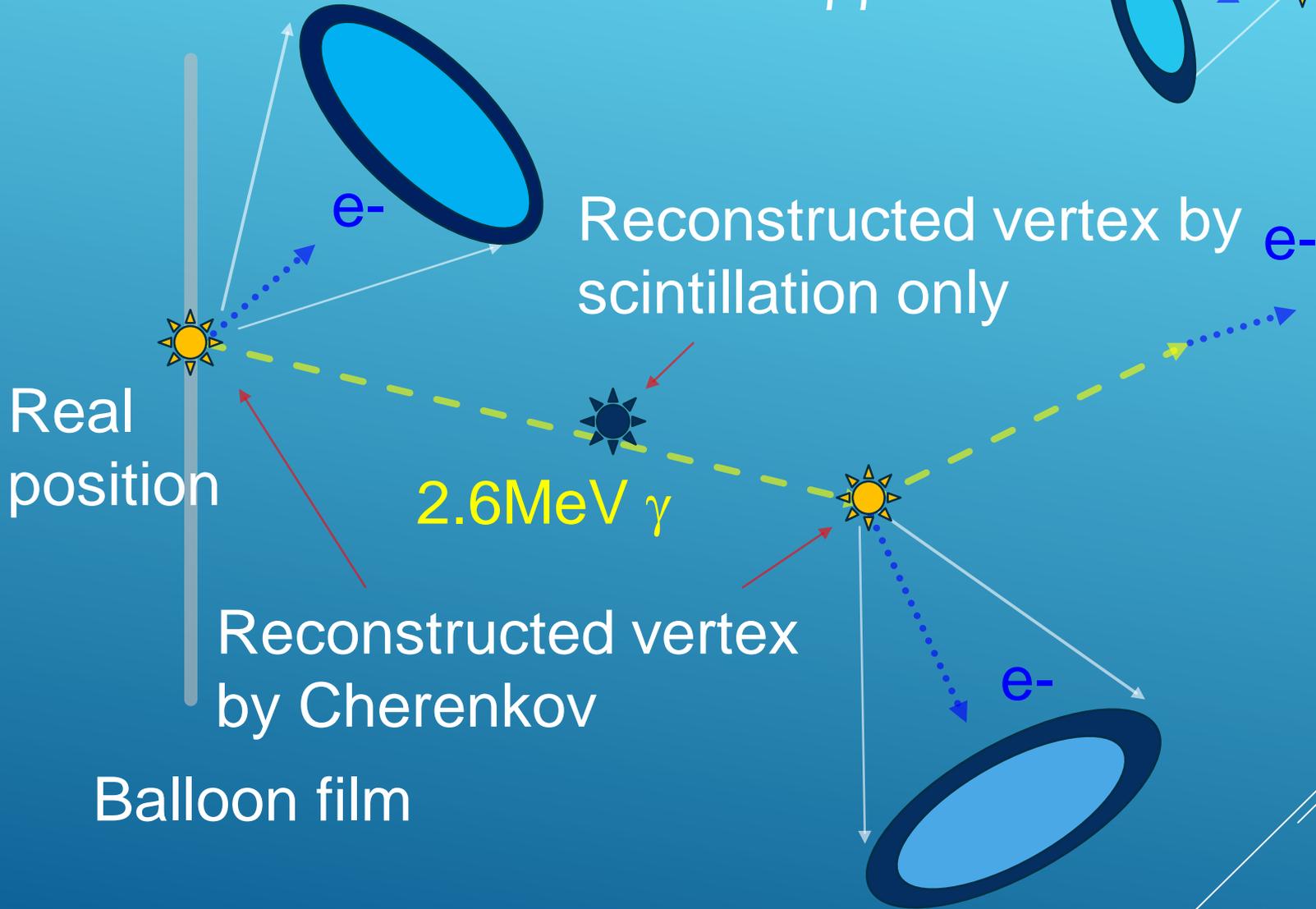
^{214}Bi : β and multi γ s (609keV, 1.12MeV)



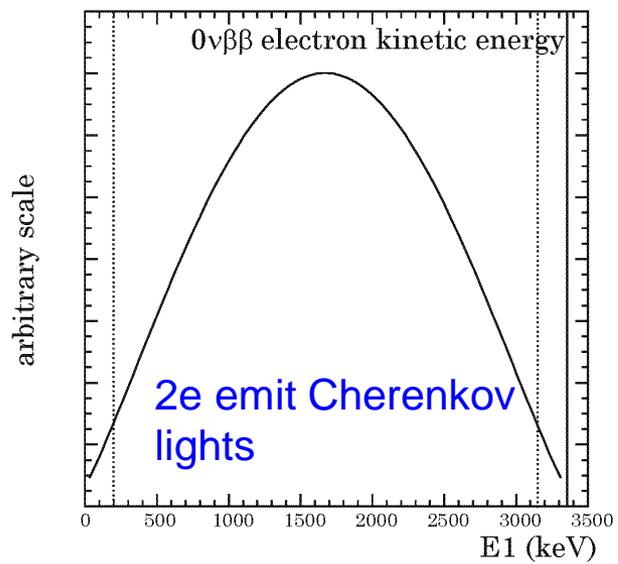
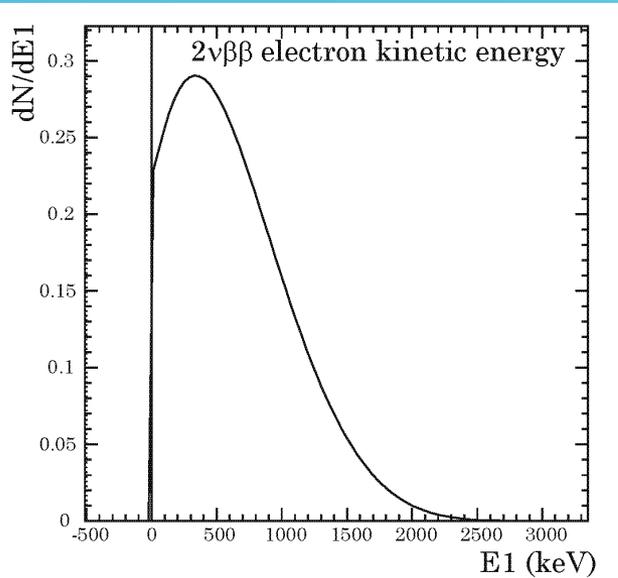
- No Cherenkov lights (scintillation only)
 - α -particle and low energy e/γ
- Observed Cherenkov lights
 - Can we check the consistency of vertex position obtained by scintillation and Cherenkov? (if we can reconstruct vertex position using Cherenkov lights.)

^{208}Tl backgrounds

$0\nu\beta\beta$ event



Electron kinetic energy spectrum



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

$\varepsilon_i = \text{sqrt}(k_i^2 + m_e^2)$: electron energy

$W_0 = Q + 2m_e$: total release energy

Q : Q value m_e : electron mass

θ : opening angle \mathcal{F} : Fermi func.

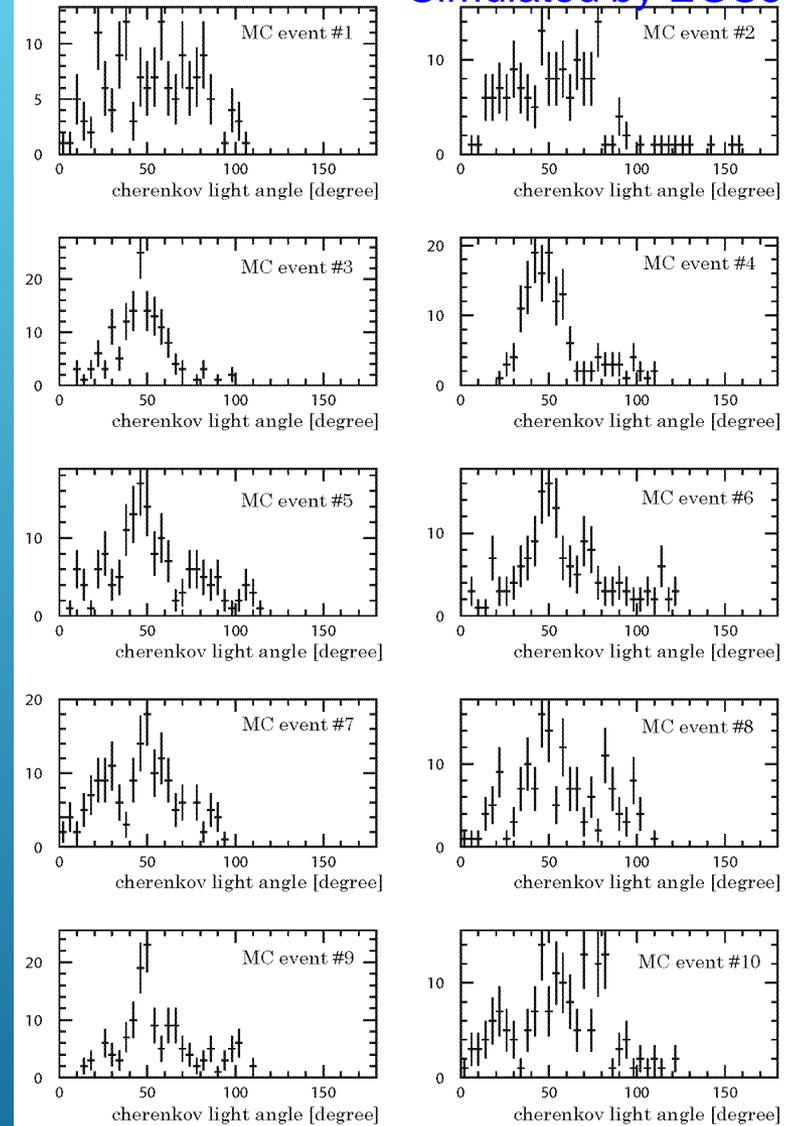
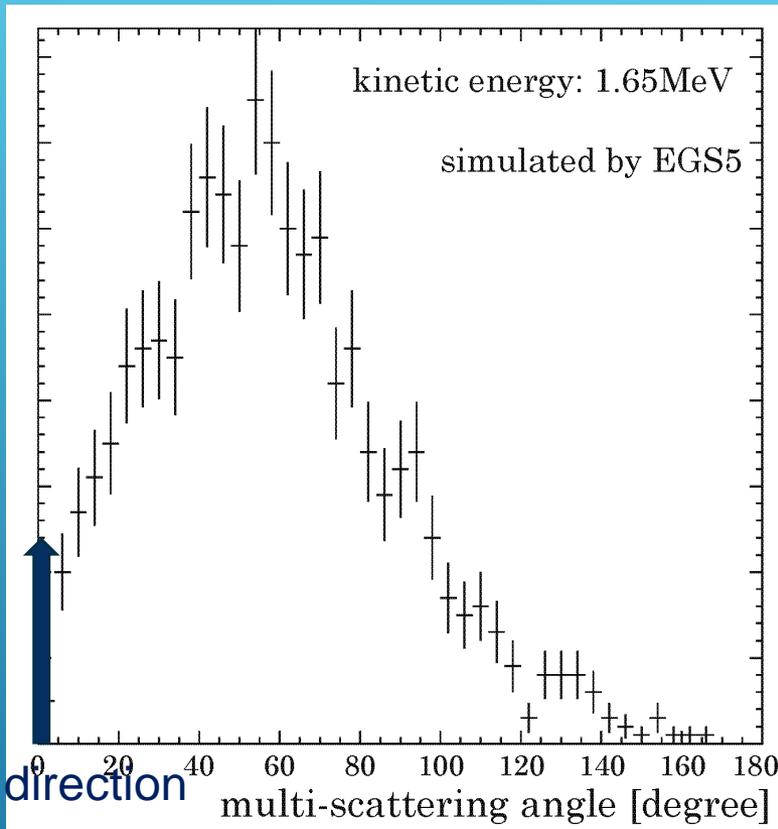
ε_i can generate independently within energy conservation.

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

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

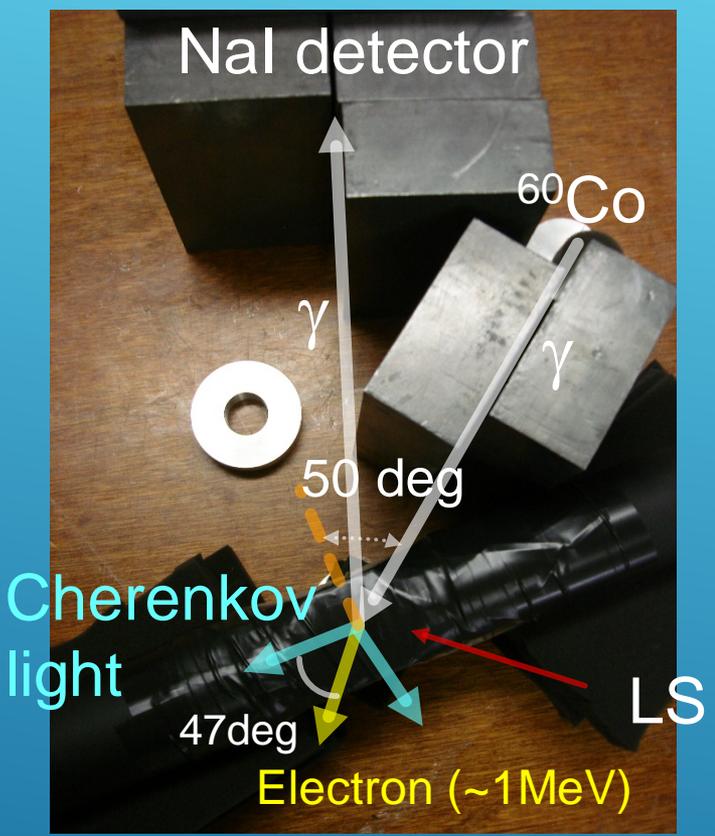
Multiple scattering of electrons

Simulated by EGS5

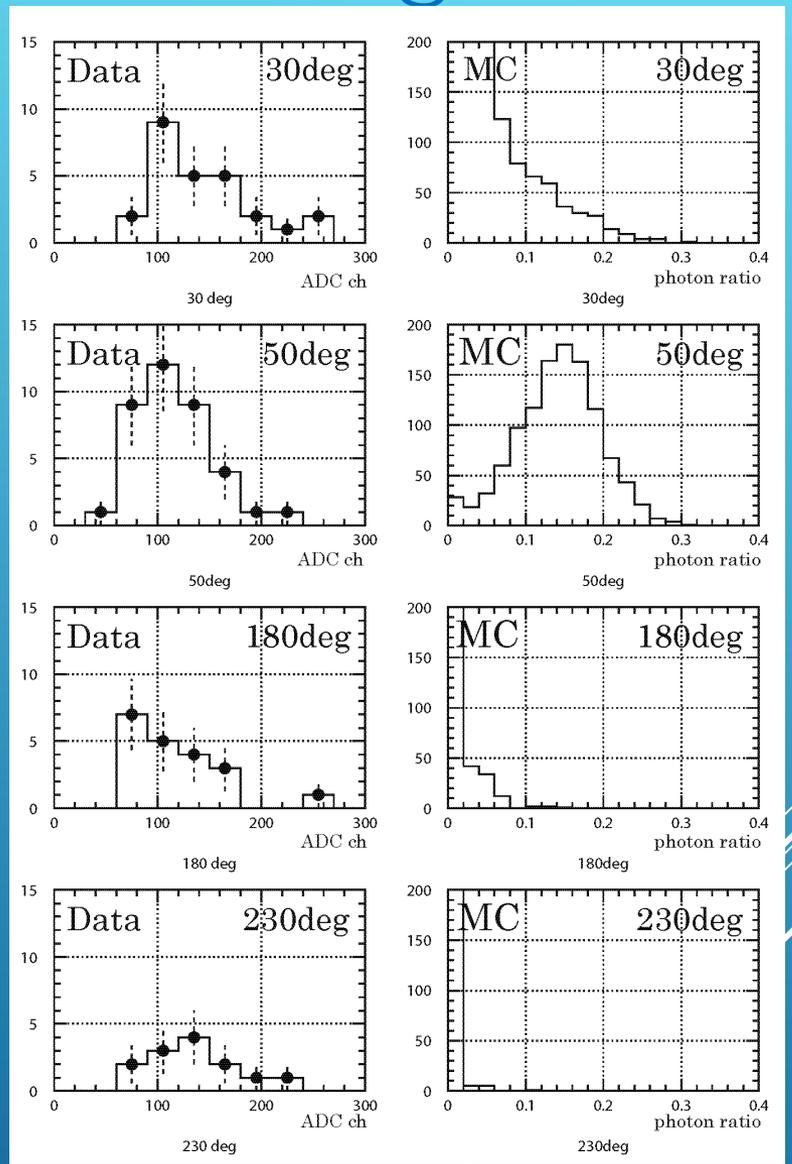


In fact, electrons should be scattered by multiple scattering, however Cherenkov light looks have **some clusters** in direction.

Measurement of Cherenkov light

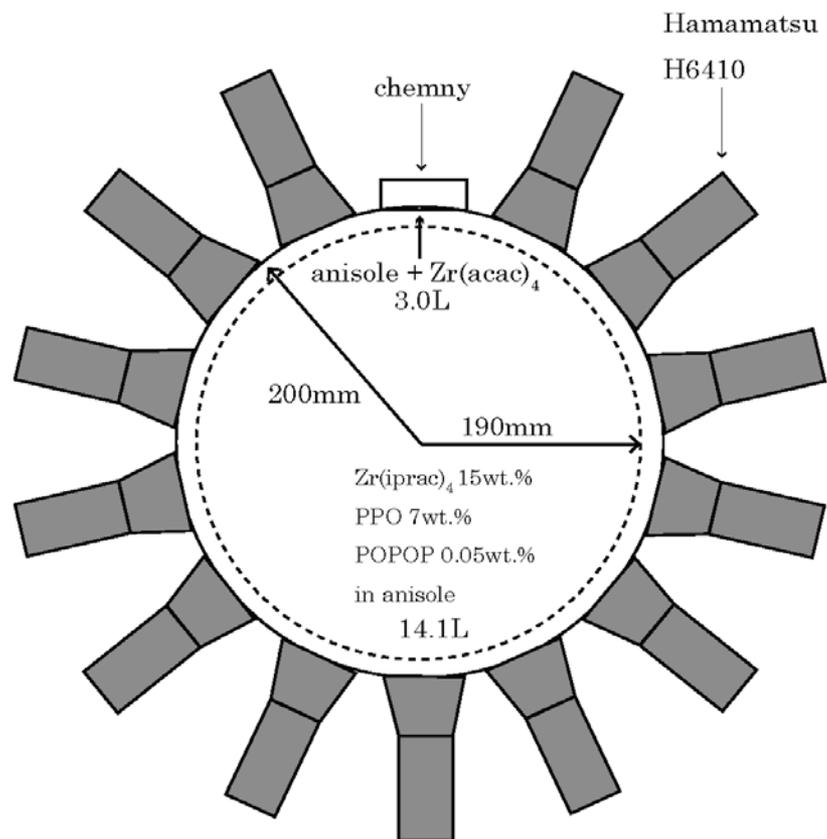


Comparison of light yields with several angle with respect to direction of incident gamma.



ZICOS proto-type detector

ZICOS proto-type detector



Total PMT: 61

Photo coverage : 40%

Scintillation (energy) + Cherenkov (BG reduction)

10⁻⁴ scale of ZICOS detector

Purpose of Performance :

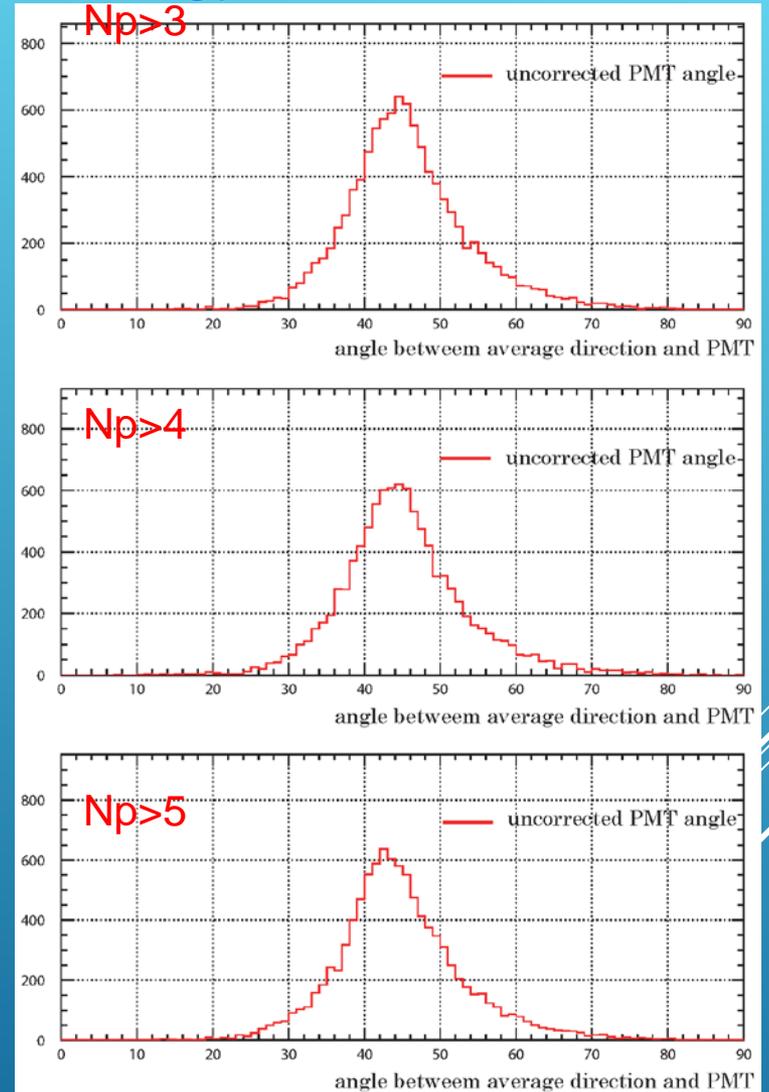
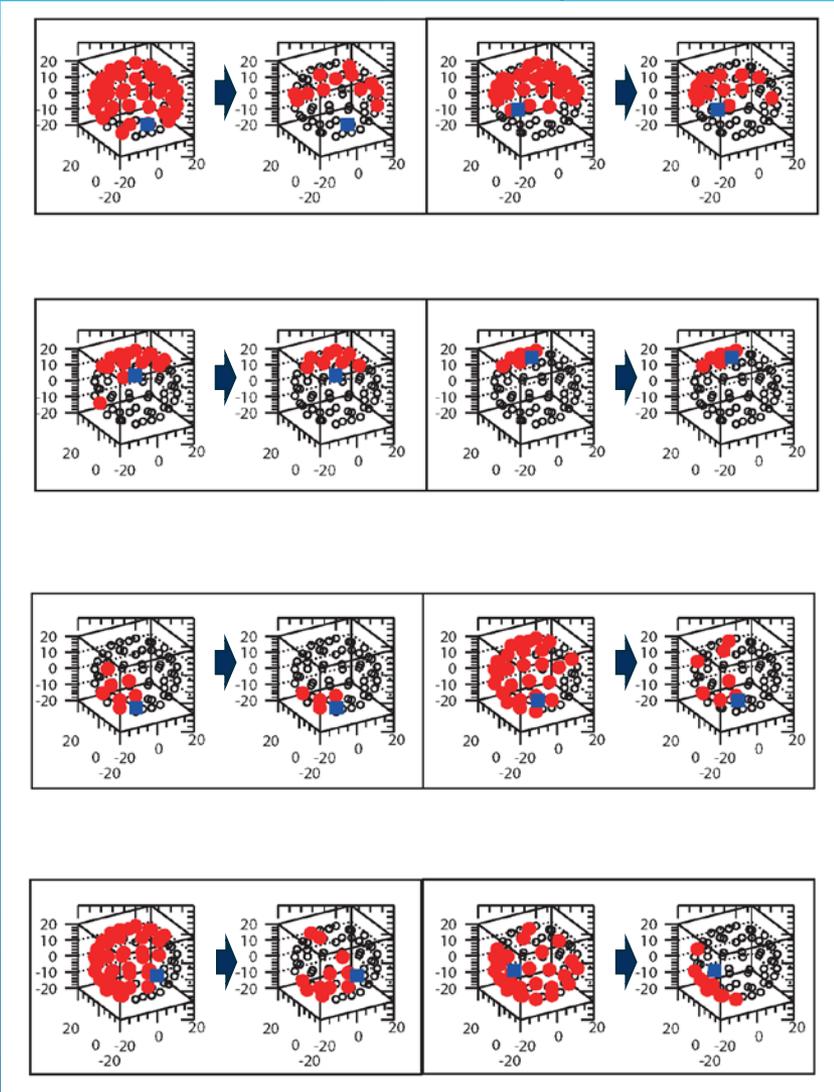
- 1) Energy resolution
- 2) BG reduction study using Cherenkov lights

Demonstration :

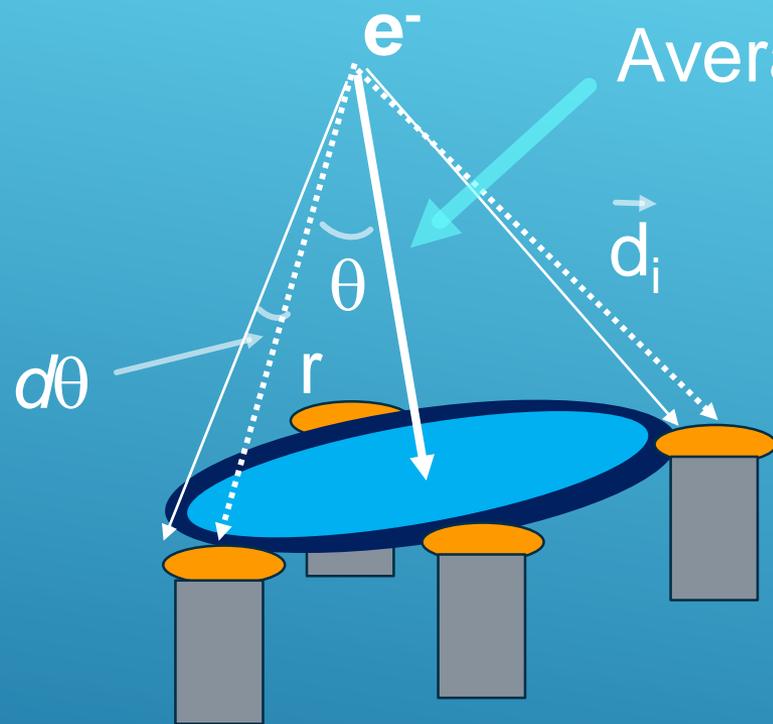
- 1) ⁹⁶Zr : 10g (same as NEMO-3) with natural abundance of Zr.
- 2) Half life limits $T_{1/2}(0\nu\beta\beta) > 1.0 \times 10^{22}$ years will be obtained, if no BG was found in 200 days' measurement.

Simulation of Hit pattern of Cherenkov

Simulated by EGS5 (kinetic energy 1.65MeV)



Correction of incident position on PMT



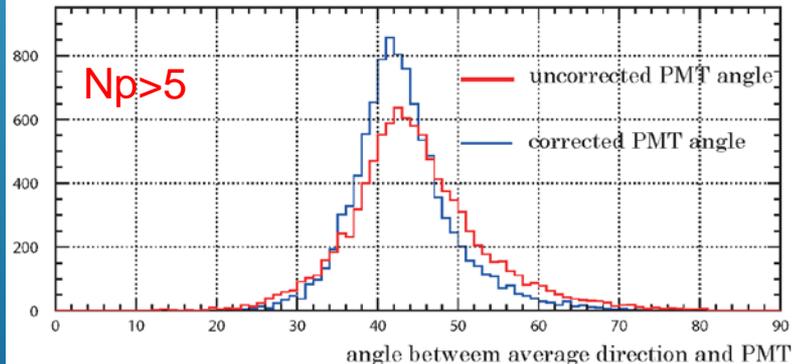
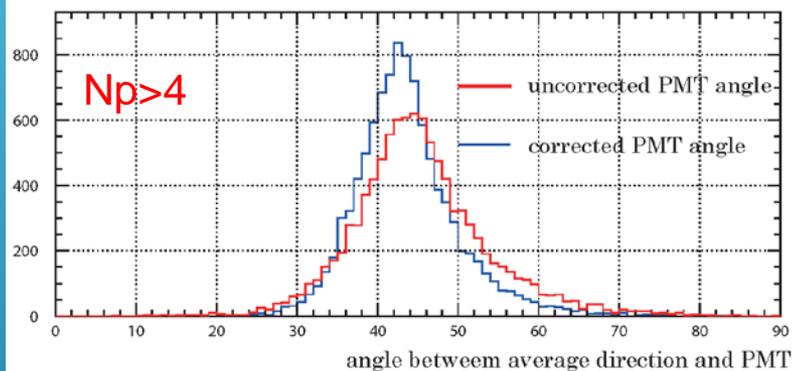
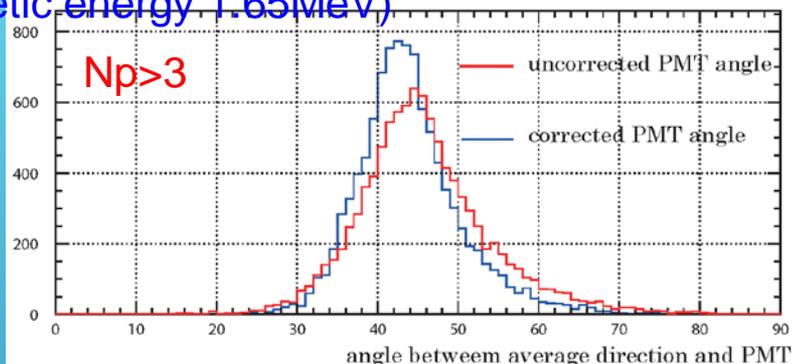
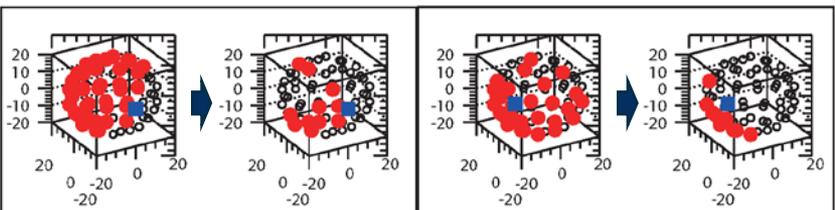
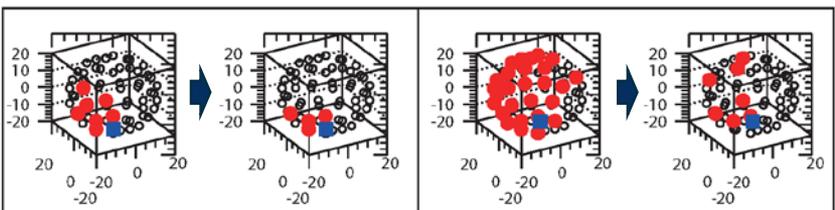
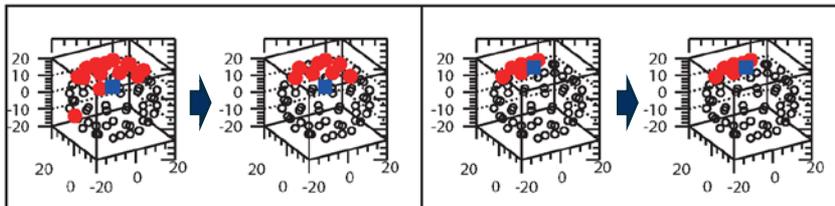
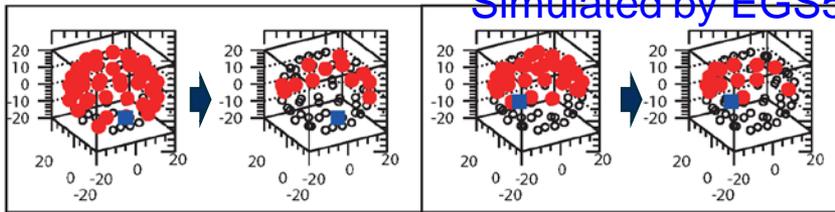
$$\tan d\theta_{\max} = (s/2)/r$$

s : radius of PMT surface
 r : distance between PMT and vertex position

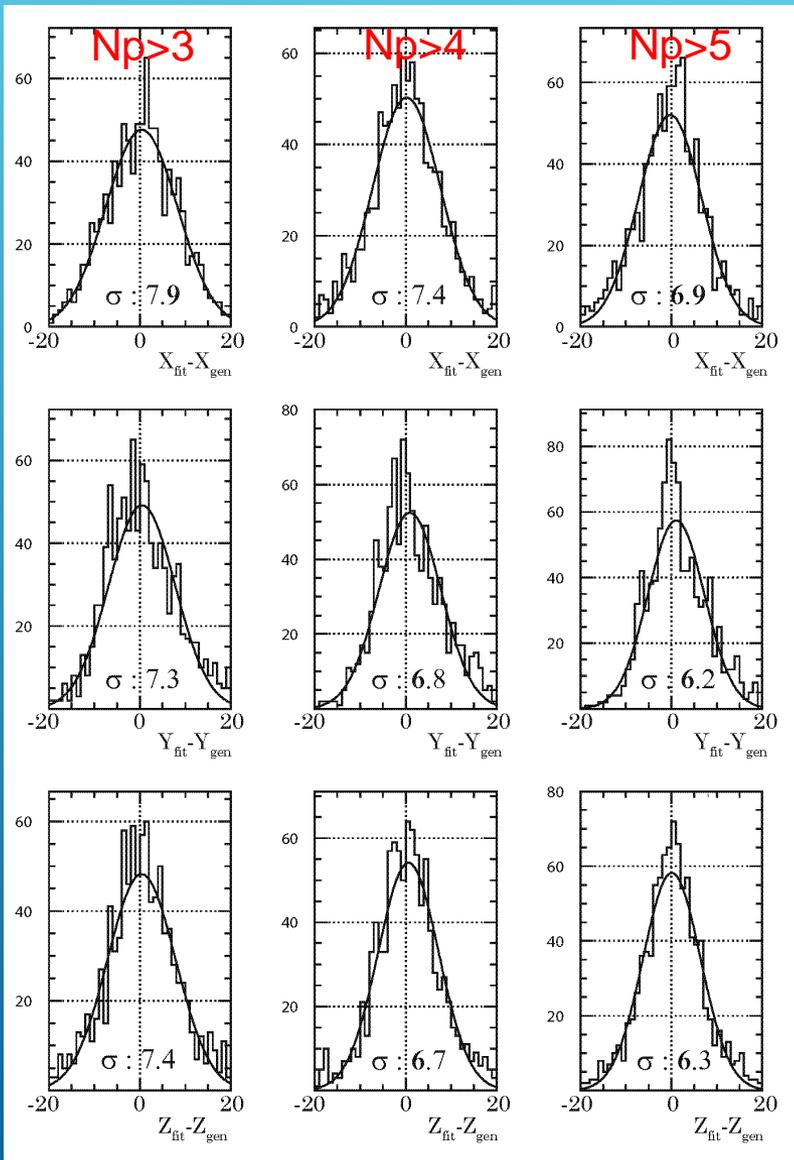
Wider or narrower angle θ could be corrected by $d\theta$.

Hit pattern after correction

Simulated by EGS5 (kinetic energy 1.65MeV)



Vertex reconstruction using chi-square



Simple vertex reconstruction using multi-photon PMTs makes clusters around generated position, even though electron's multiple scattering .

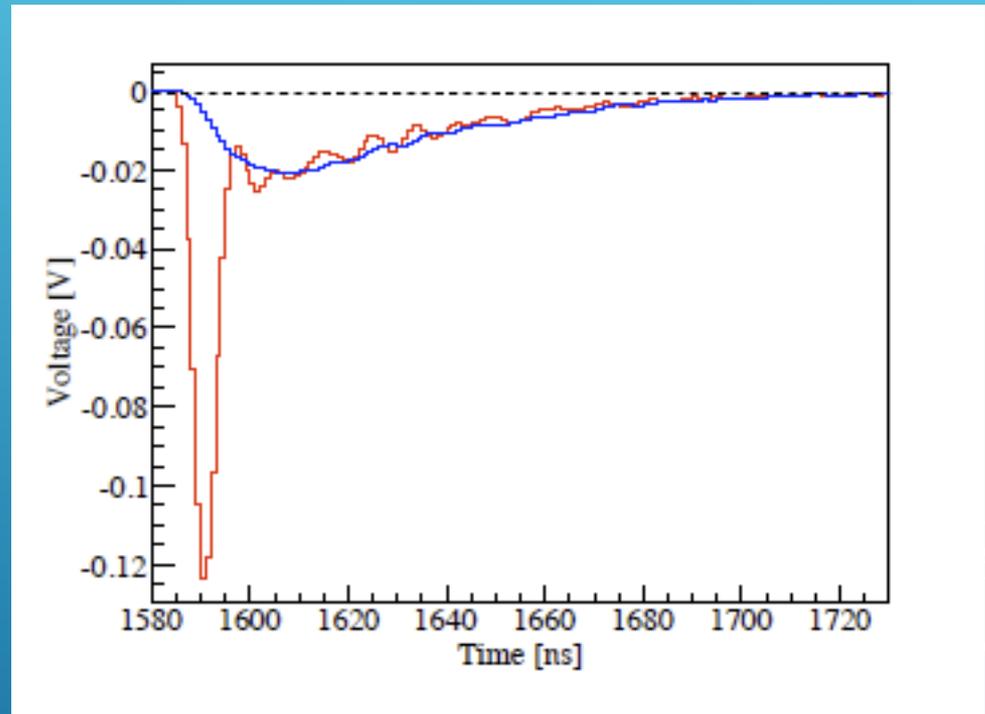
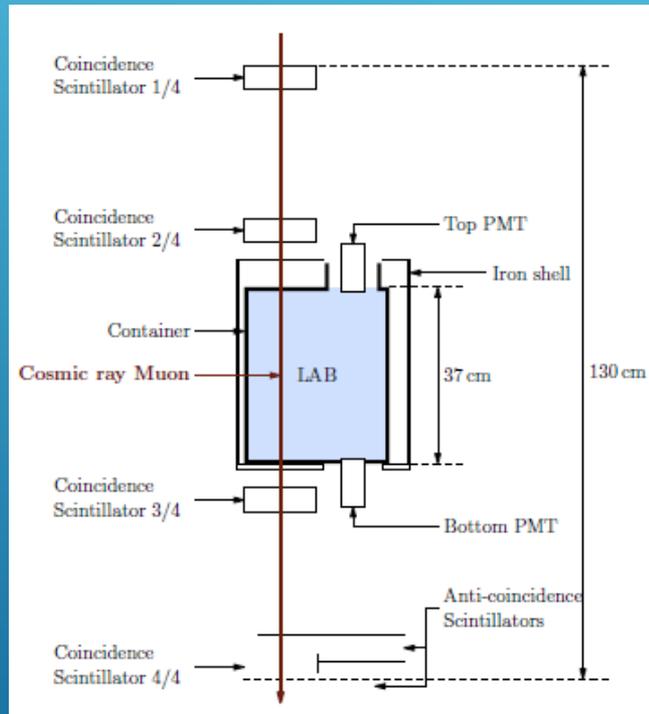


Real (not proto-type) ZICOS detector will use not only hit pattern, but also **light arrival time** in order to reconstruct vertex correctly , so that much better vertex reconstruction resolution should be expected.

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.

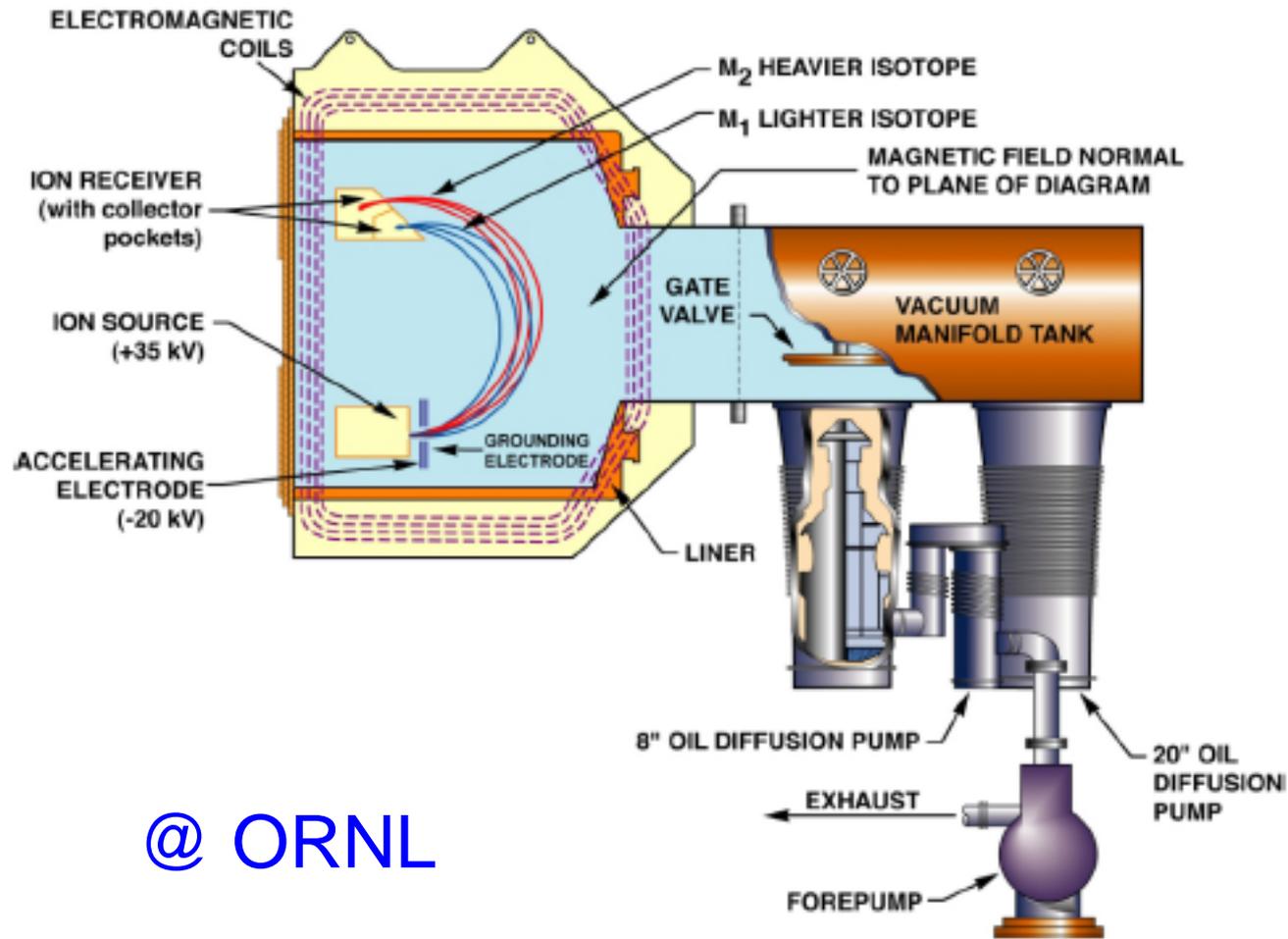
Summary

- ▶ Liquid scintillator containing Zr(iprac)_4 has been developed for ZICOS experiment.
- ▶ Succeeded to obtain Liquid scintillator with 10 wt.% concentration of Zr(iprac)_4 has $48.7 \pm 7.1\%$ for light yield to BC505 and $3.5 \pm 0.5\%$ at 3.35MeV (assuming 40% photo coverage) for energy resolution.
- ▶ We are developing new technique for a background reduction using Cherenkov light for vertex reconstruction. (available for LS family like KL-Zen, SNO+, etc)
- ▶ ZICOS sensitivity : $T_{1/2}^{0\nu} > \sim 2 \times 10^{26} \text{y}$; and $\langle m_\nu \rangle < 0.04\text{-}0.06 \text{eV}$ (QRPA) if both 58.5% enrichment and BG level at $1/20 \times \text{KamLAND-Zen}$ achieved.
- ▶ We will try also Nd(iprac)_3 because larger Q-value (3.67MeV) and twice larger natural abundance (6%).
- ▶ We are now open for new collaborators.

BACKUP

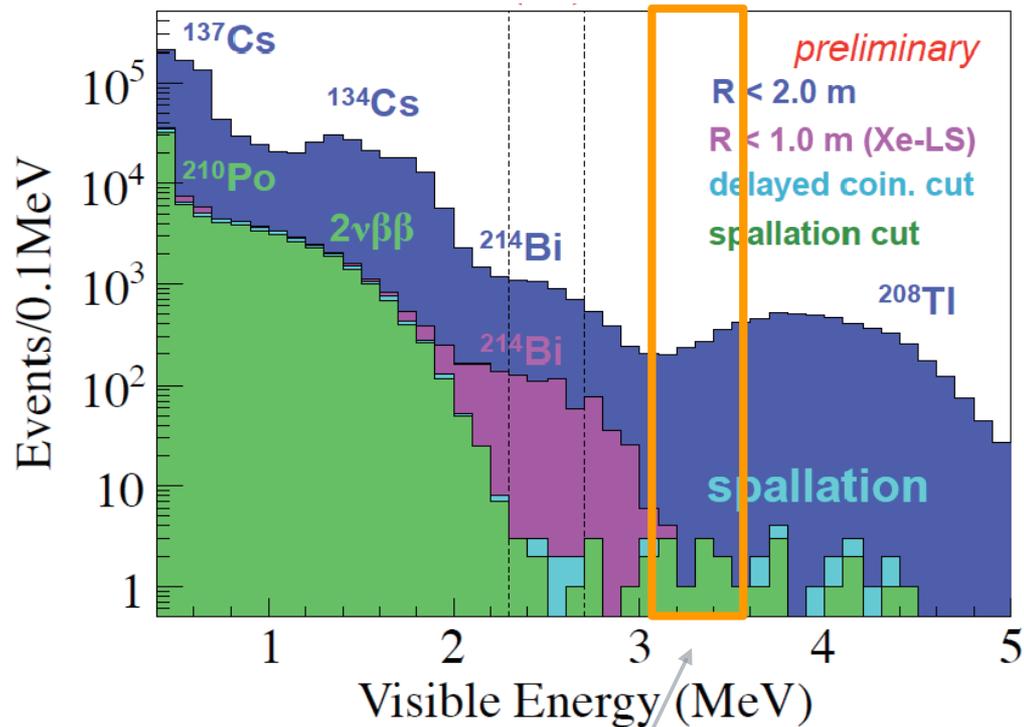
Electromagnetic isotope separation

Schematic of a Calutron



@ ORNL

Backgrounds around 3.35MeV



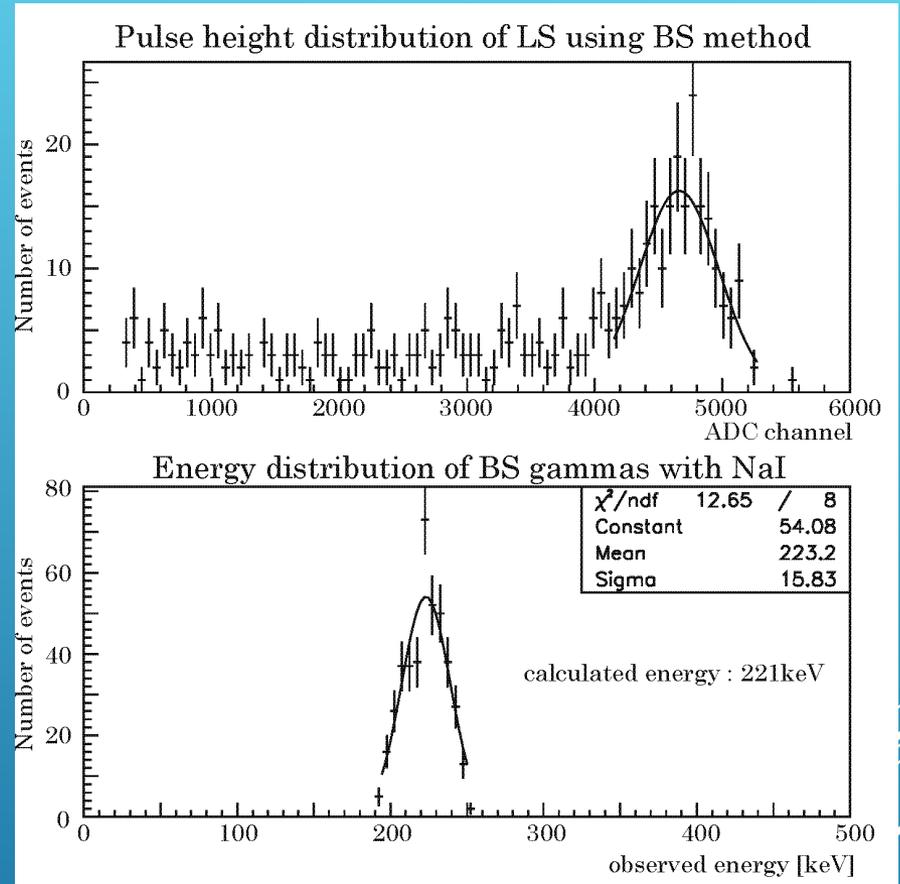
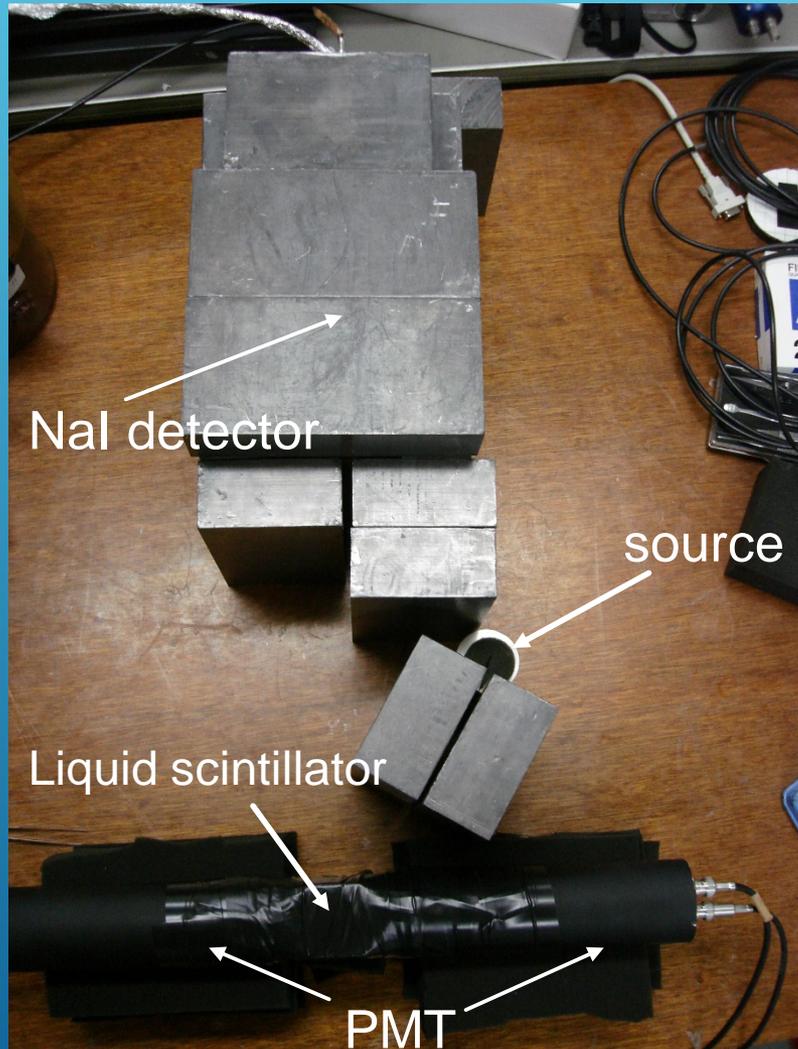
I.Shimizu@Neutrino2014

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

^{214}Bi contaminates within 1m inner volume, but most of events could be removed by Bi-Po tagging.

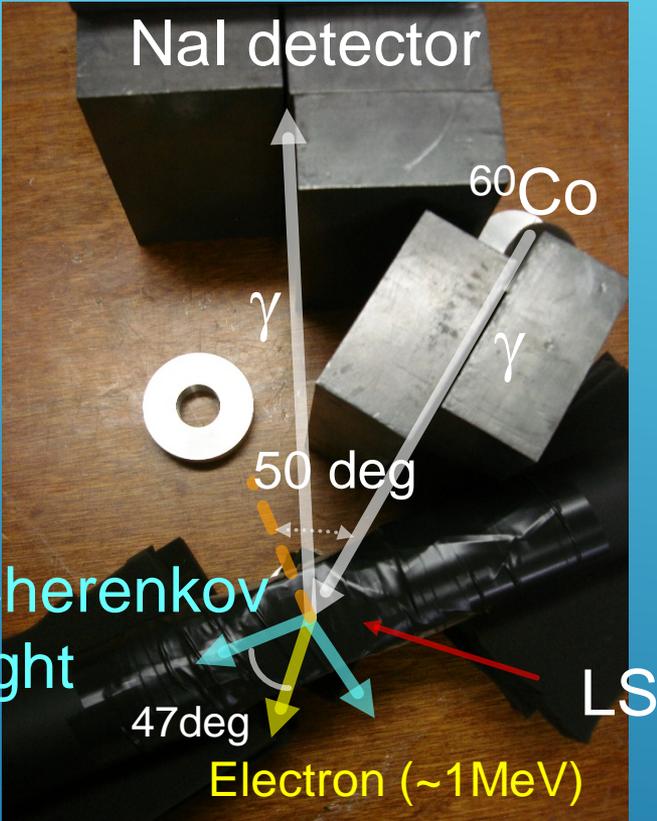
If α particle may be absorbed in films, we can not use tagging.

Backscattering method



Single peak could be used even in liquid scintillator.

Measurement of Cherenkov light

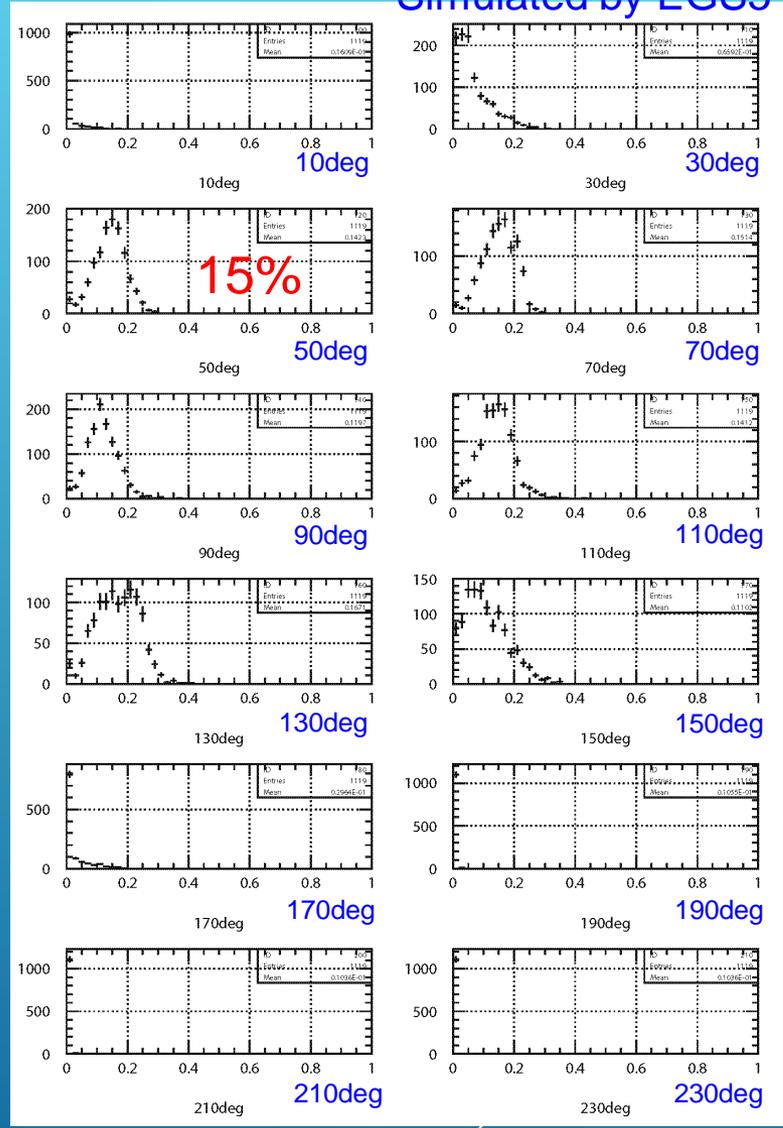


Cherenkov light

Electron (~1MeV)

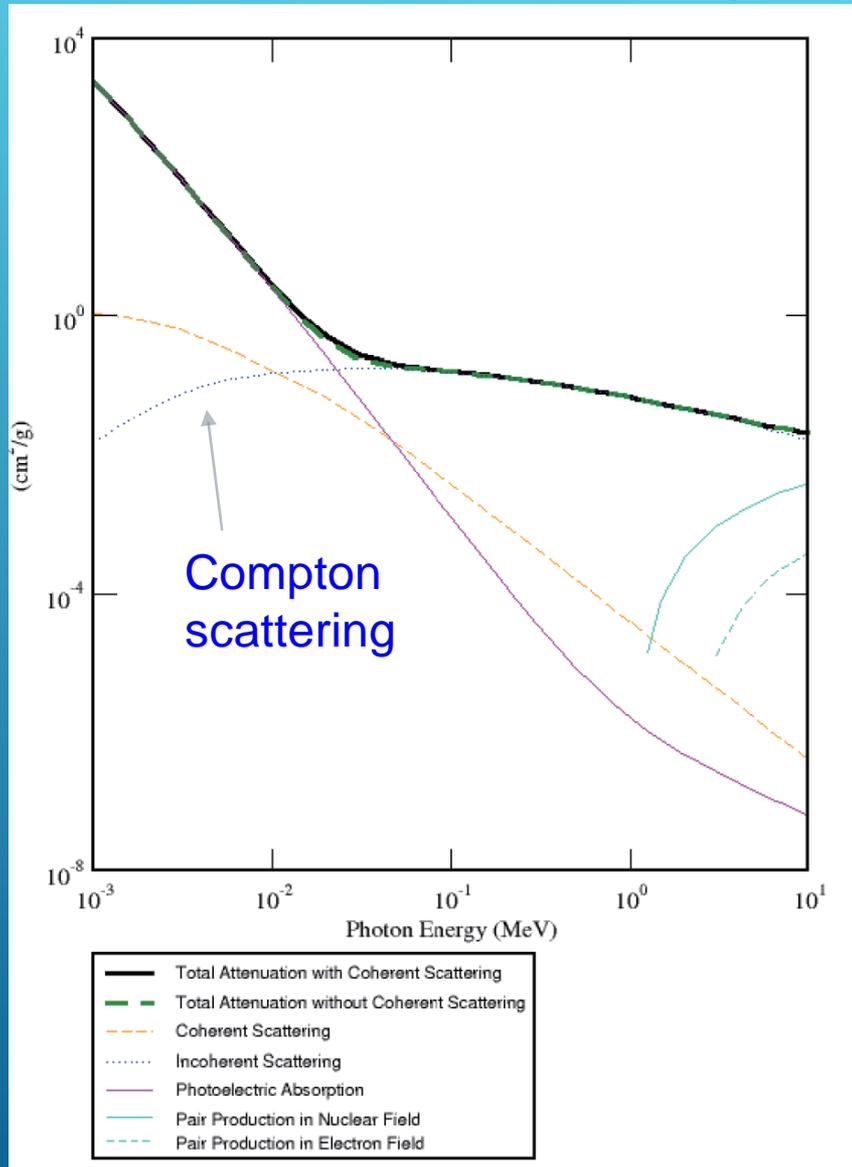
LS

Simulated by EGS5



Comparison of light yields with or without SC-37 filter using back scattering method.

Cross section of gamma in anisole

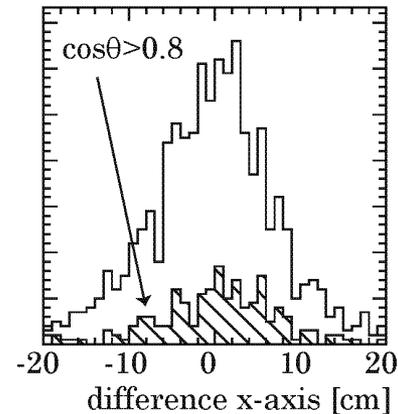
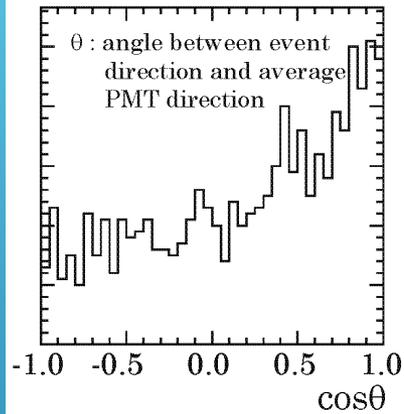


Most of few MeV range γ s interact with anisole via Compton scattering.

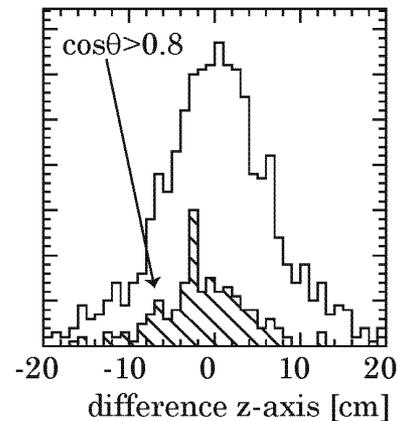
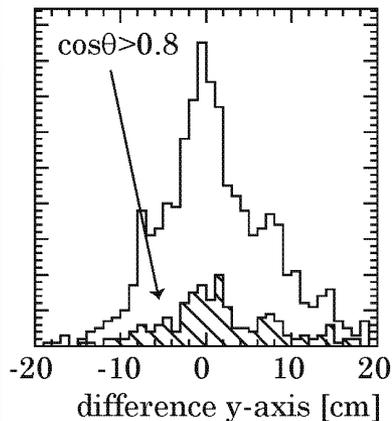


At least, one event could have Cherenkov hits.

Event direction and average PMT direction

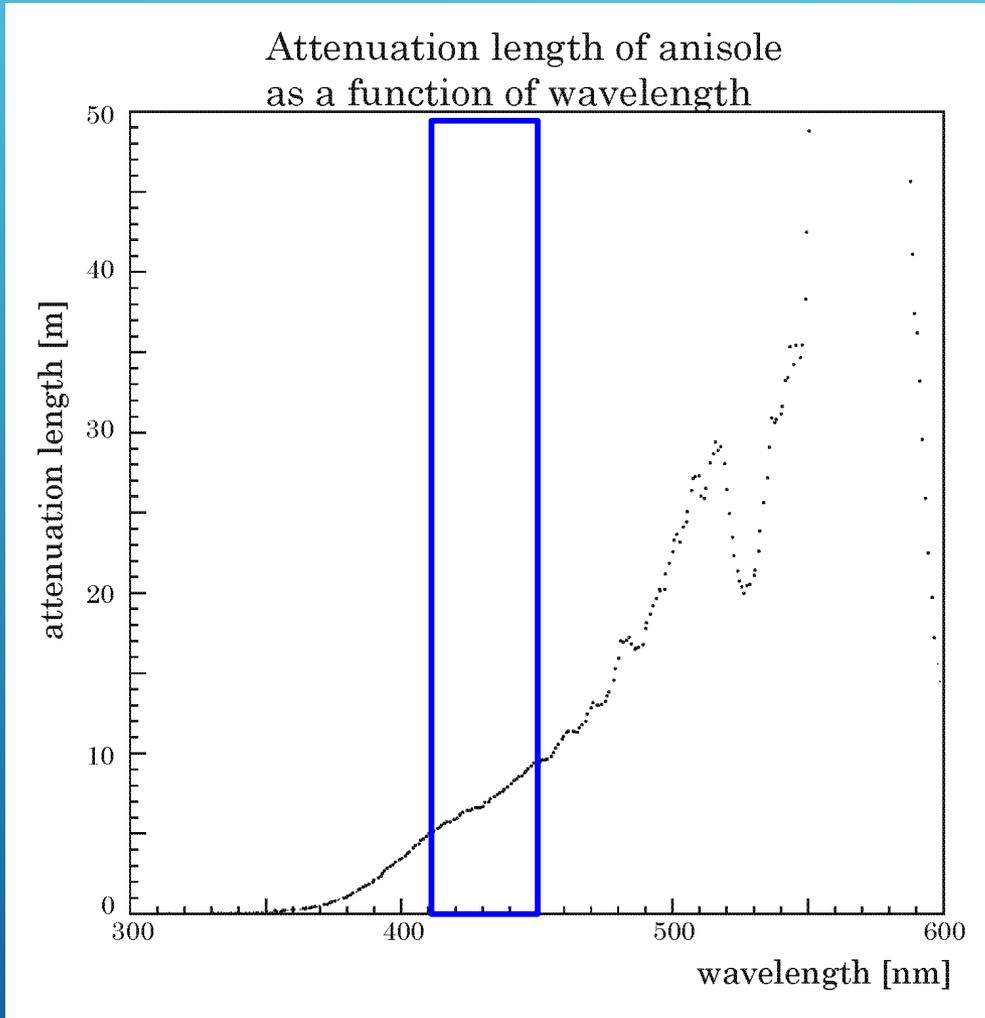


No relation between event direction and averaged PMT direction was found.



Clustered PMTs should be made by longest track within multiple scattering.

ATTENUATION LENGTH OF ANISOLE

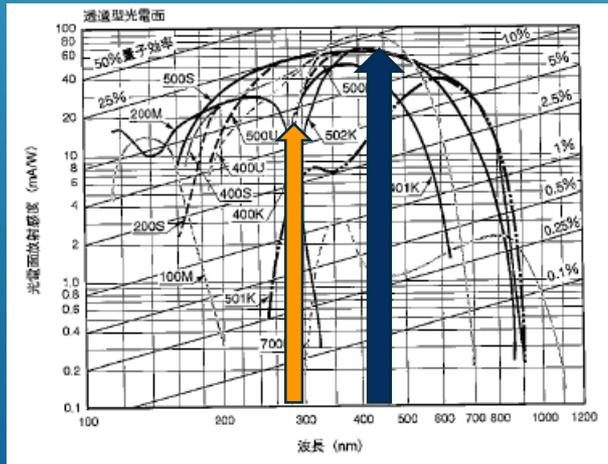
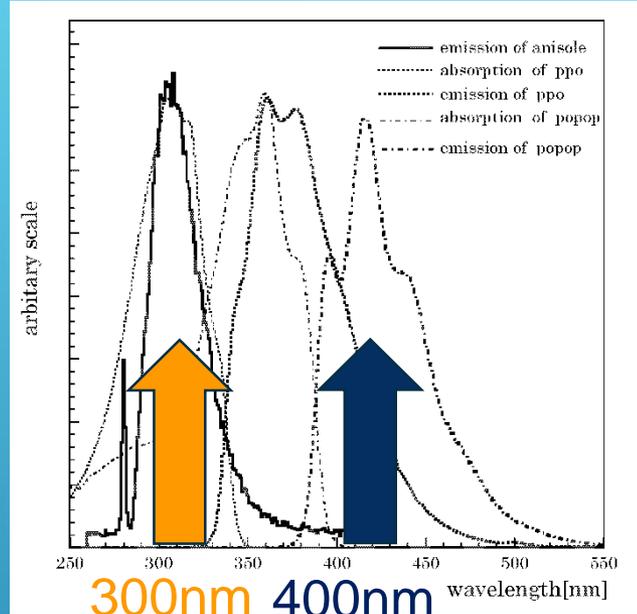
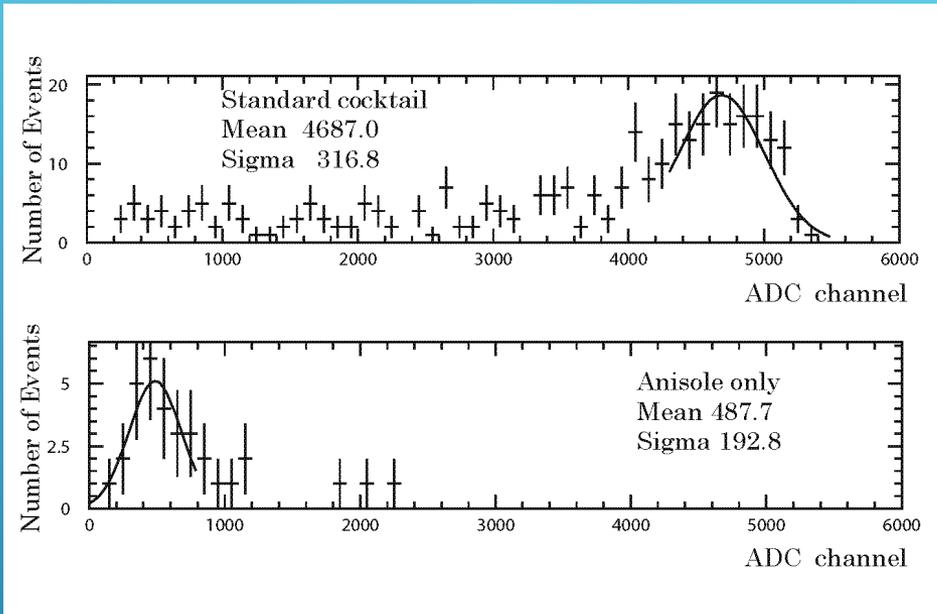


Attenuation length of scintillation light from POPOP (~450nm) was obtained as ~6m.



No problem for radius of ZICOS detector.

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)

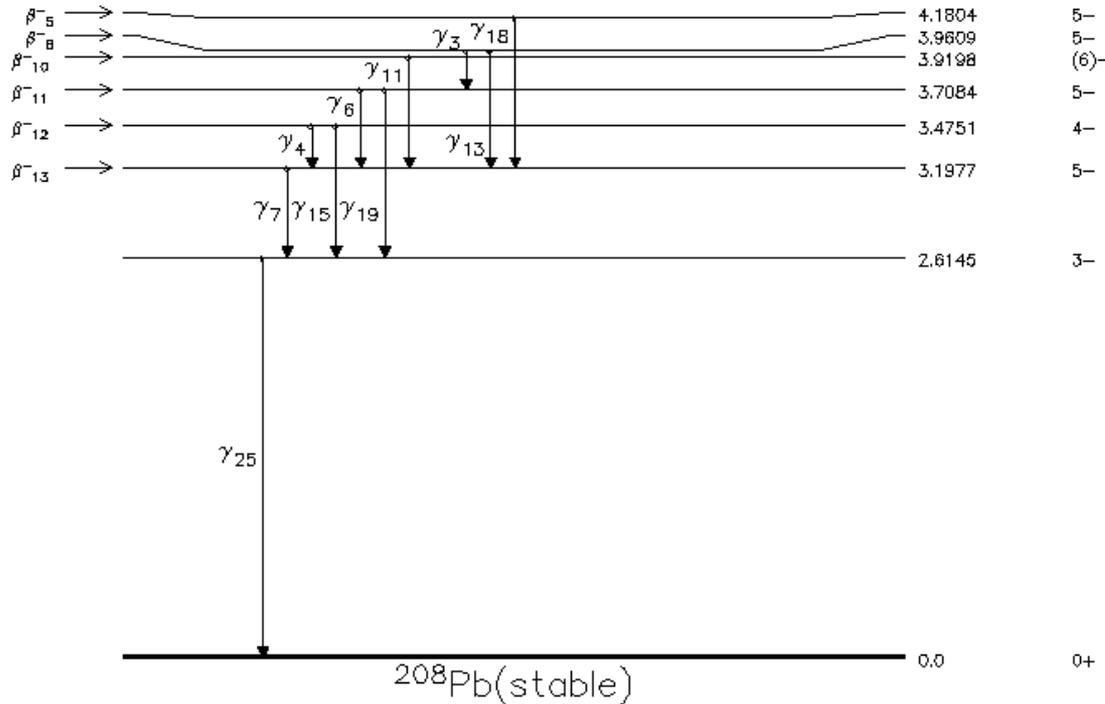
Thallium-208 radiation branch

^{208}Tl (3.053 m)

5(+)
0.0

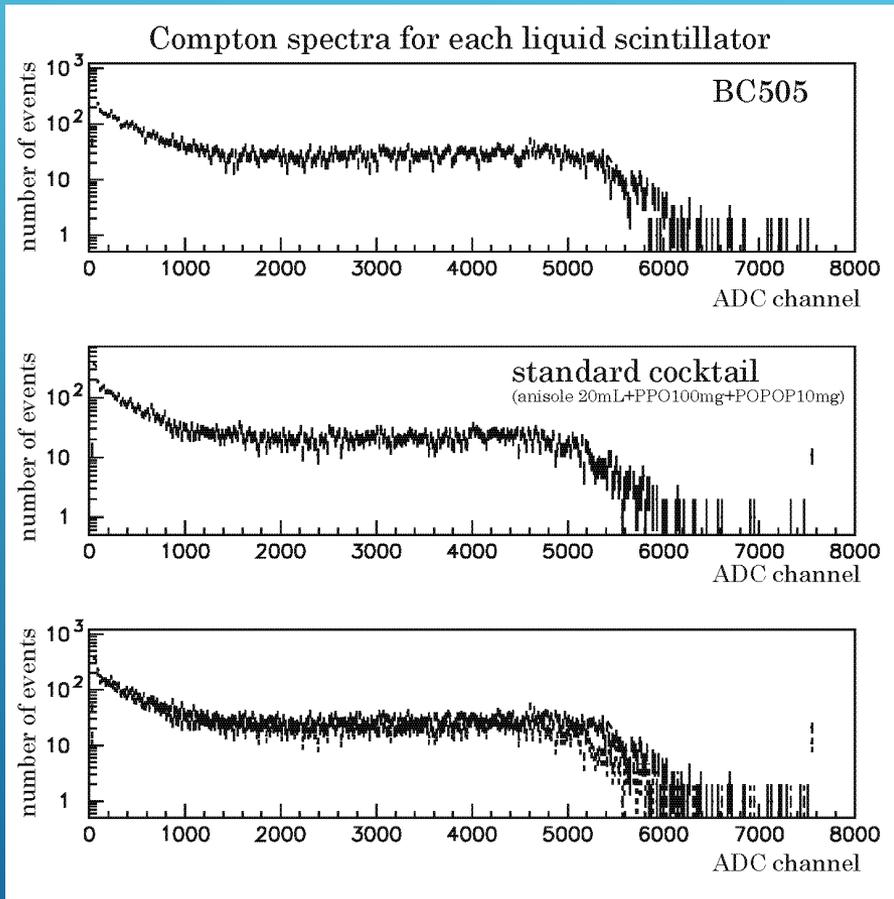
β^- decay 100.00 %

$Q_{\beta^-} = 4.9940$



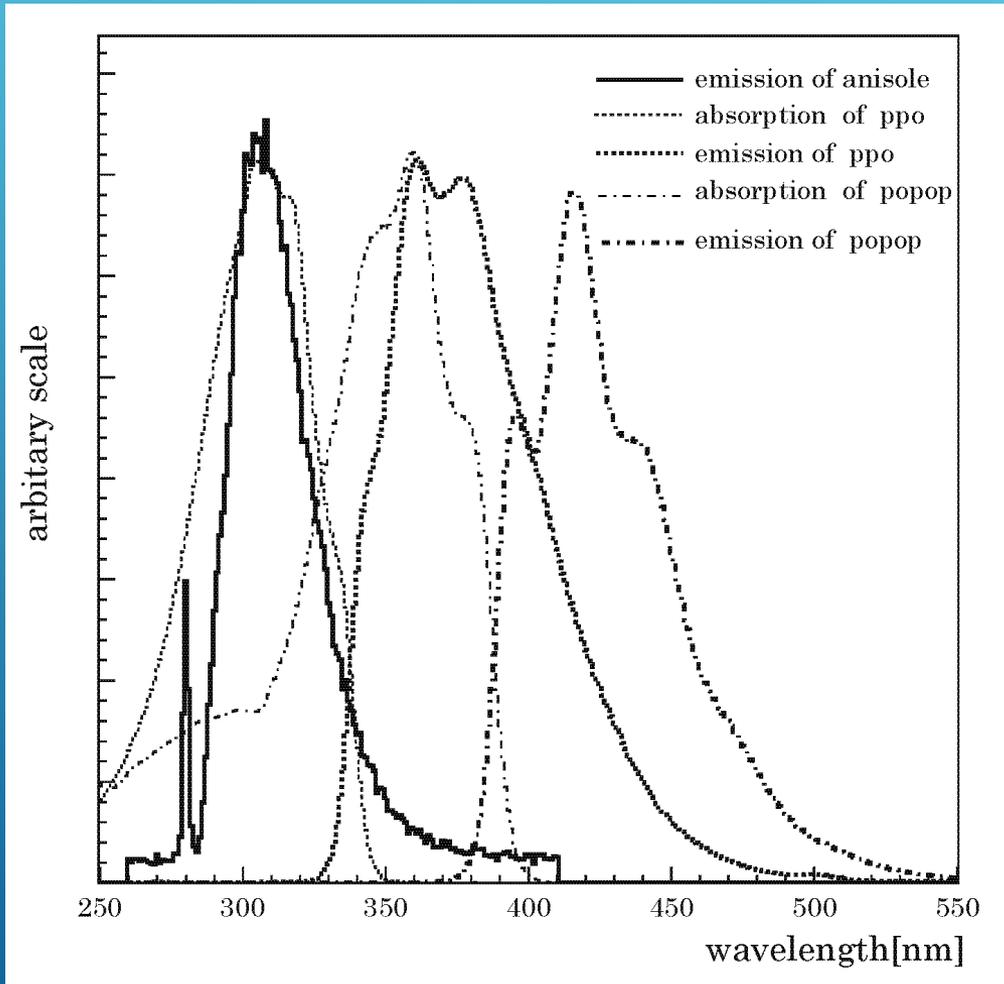
Radiations	$y(i)$ (Bq-s) ⁻¹
beta- 5	2.27×10^{-03}
beta- 8	3.09×10^{-02}
beta- 10	6.30×10^{-03}
beta- 11	2.45×10^{-01}
beta- 12	2.18×10^{-01}
beta- 13	4.87×10^{-01}
ce-K, gamma 3	4.04×10^{-03}
gamma 4	6.31×10^{-02}
ce-K, gamma 4	2.84×10^{-02}
ce-L, gamma 4	4.87×10^{-03}
gamma 6	2.26×10^{-01}
ce-K, gamma 6	1.97×10^{-02}
ce-L, gamma 6	3.32×10^{-03}
gamma 7	8.45×10^{-01}
ce-K, gamma 7	1.28×10^{-02}
ce-L, gamma 7	3.51×10^{-03}
gamma 13	1.81×10^{-02}
gamma 15	1.24×10^{-01}
ce-K, gamma 15	2.80×10^{-03}
gamma 19	3.97×10^{-03}
gamma 25	9.92×10^{-01}

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.

Emission and absorption spectra for solvent and solute in standard cocktail

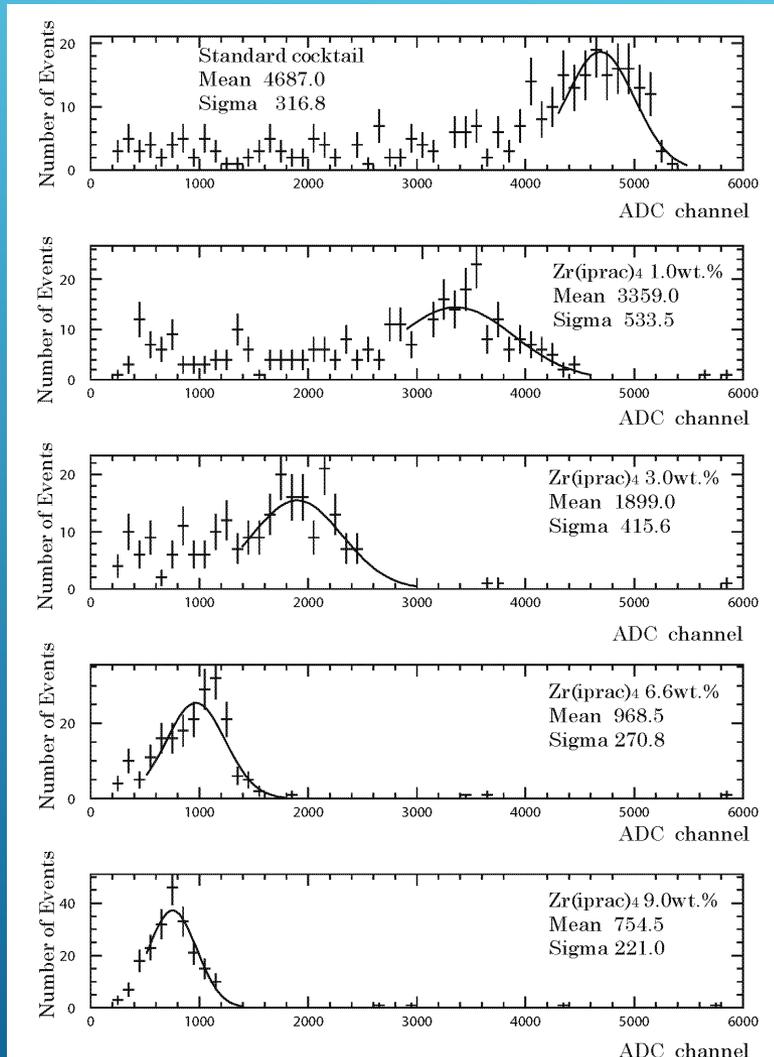


PPO absorbed most of emission lights from anisole.



Effectively the energy was transferred to the secondary scintillator.

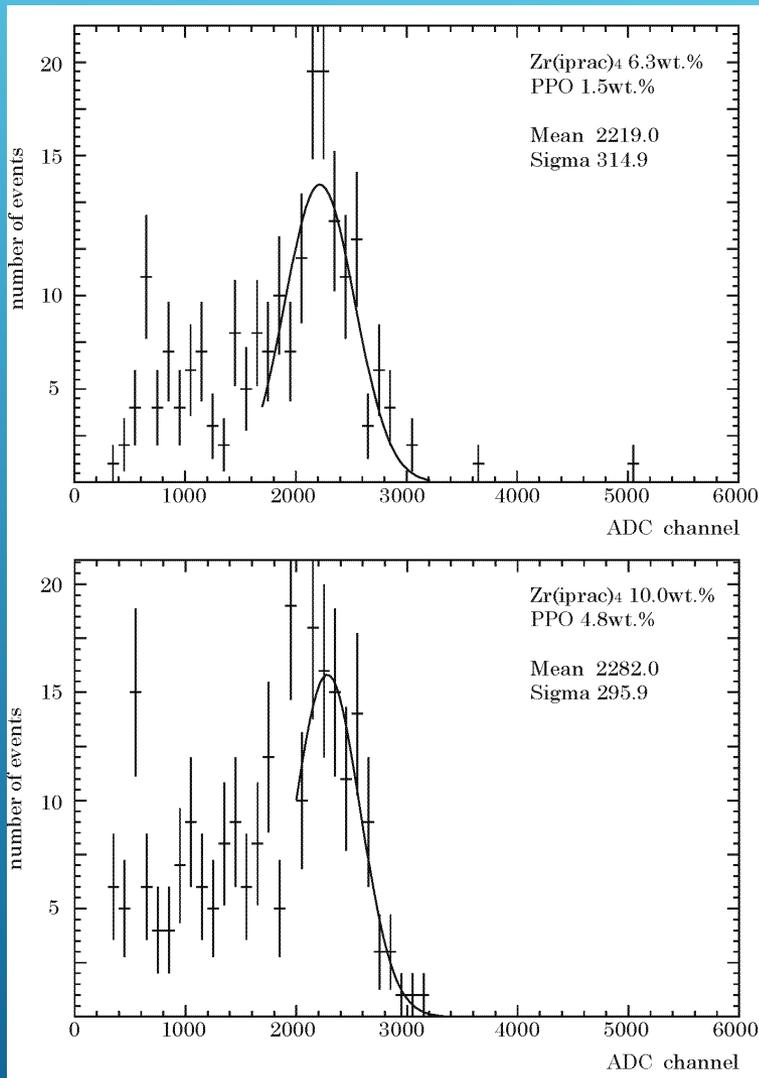
ENERGY SPECTRA FOR SEVERAL CONCENTRATION OF Zr(iprac)₄



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.

RECOVERY FOR ABILITY OF LIGHT YIELD AND ENERGY RESOLUTION

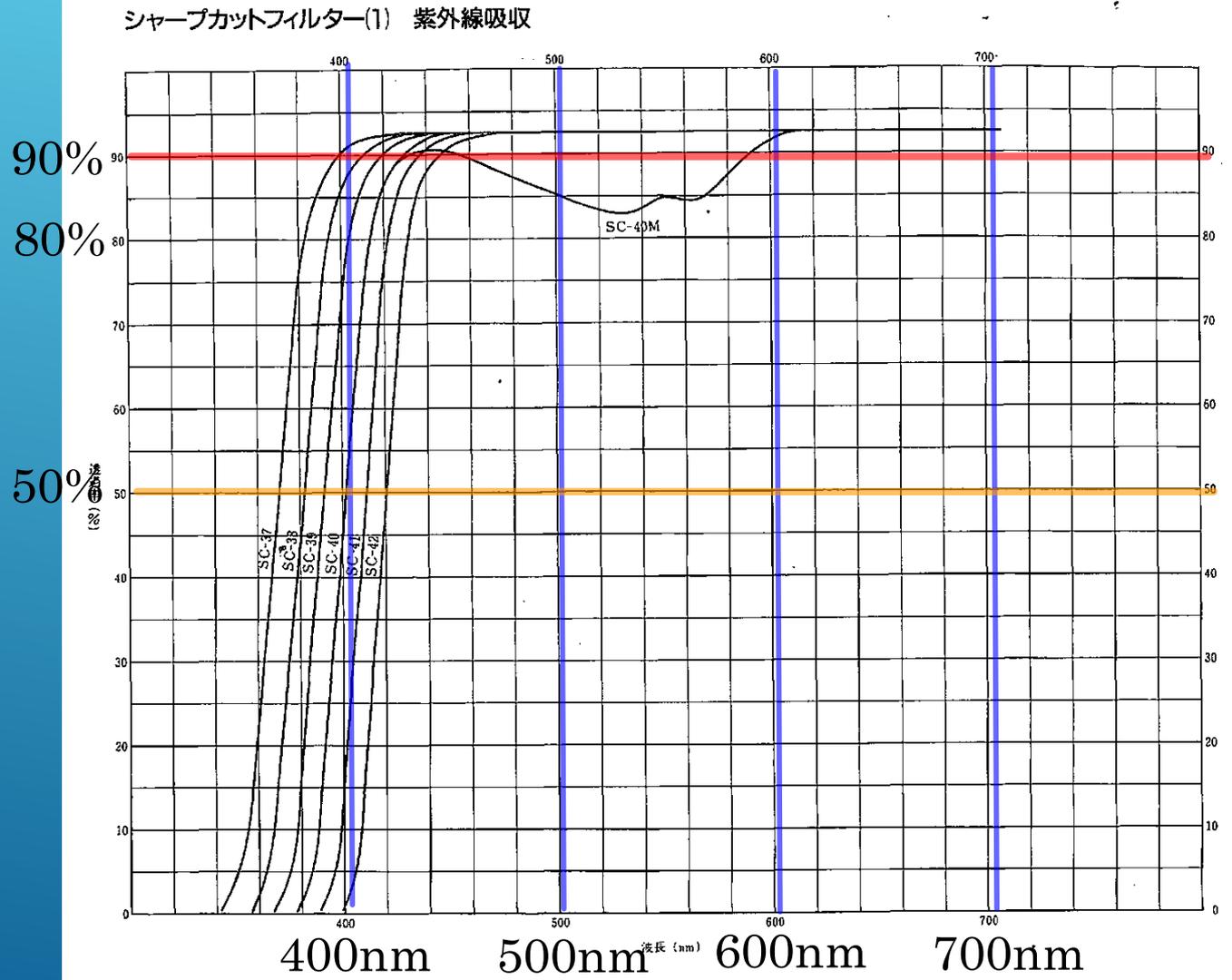


PPO helps recovering the light yield and the energy resolution.



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

UV sharp cut filter (Fuji films)



Physical constants of Liquid Scintillator

Physical Constants of SGC Liquid Scintillators

Scintillator	Light Output % Anthracene ¹	Wavelength of Maximum Emission, nm	Decay Constant, ns	H:C Ratio	Loading Element	Density	Flash Point °C
BC-501A	78	425	3.2 ¹	1.212		0.87	26
BC-505	80	425	2.5	1.331		0.877	48
BC-509	20	425	3.1	.0035	F	1.61	10
BC-517L	39	425	2	2.01		0.86	102
BC-517H	52	425	2	1.89		0.86	81
BC-517P	28	425	2.2	2.05		0.85	115
BC-517S	66	425	2	1.70		0.87	53
BC-519	60	425	4	1.73		0.87	63
BC-521	60	425	4	1.31	Gd (to 1%)	0.89	44
BC-523	65	425	3.7	1.74	Nat. ¹⁰ B (5%)	0.916	-8
BC-523A	65	425	3.7	1.67	Enr. ¹⁰ B (5%)	0.916	-8
BC-525	55	425	3.8	1.56	Gd (to 1%)	0.88	91
BC-531	59	425	3.5	1.63		0.87	93
BC-533	51	425	3	1.96		0.80	65
BC-537	61	425	2.8	0.99 (D:C)	² H	0.954	-11

* 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) : 4×10^4 photon/MeV  LY of BC505 : 1.2×10^4 photon/MeV

Natural radiative U/Th decay chain

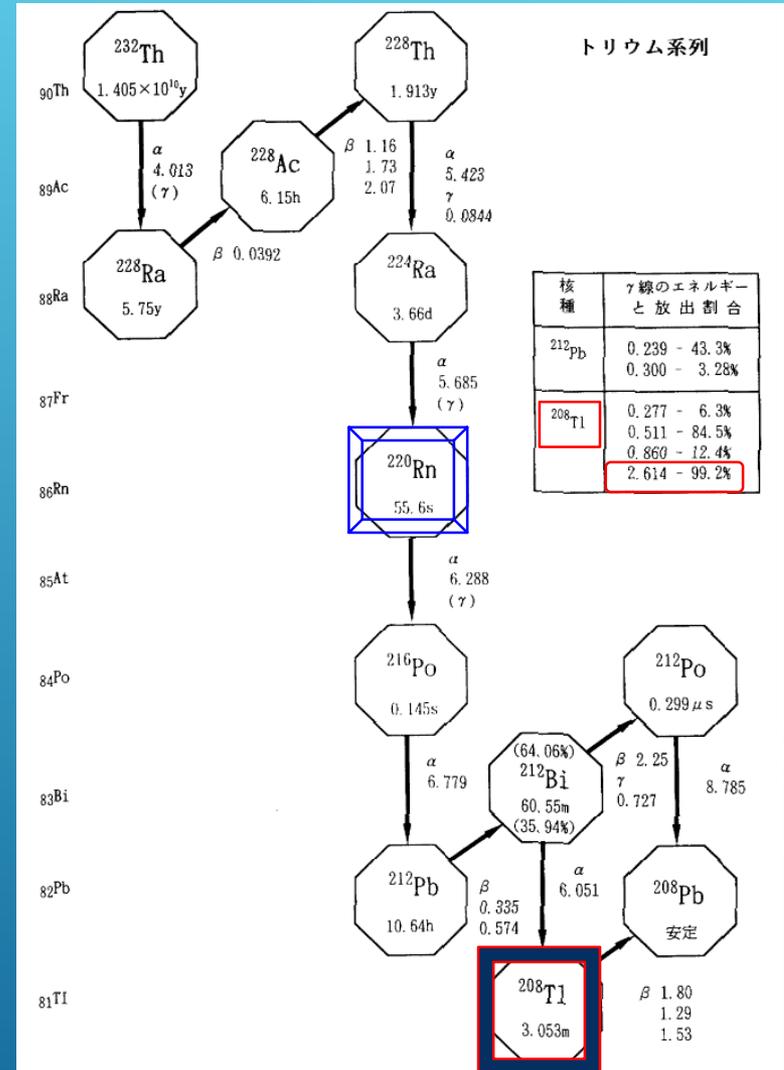
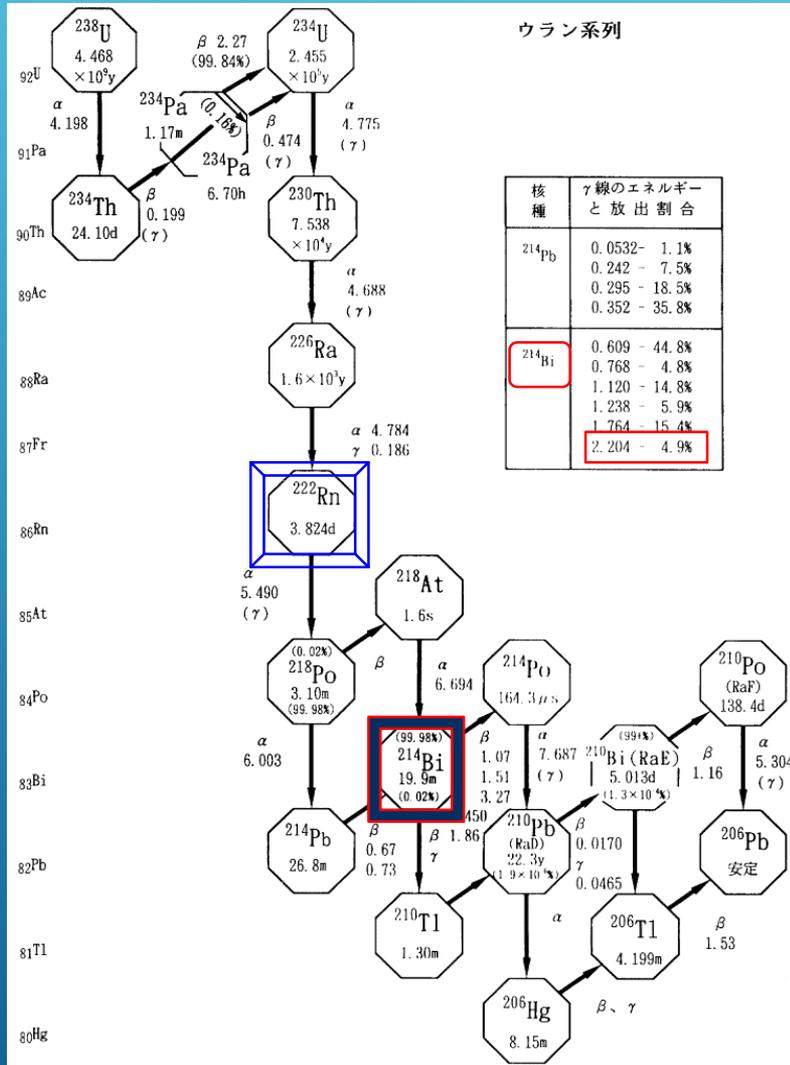
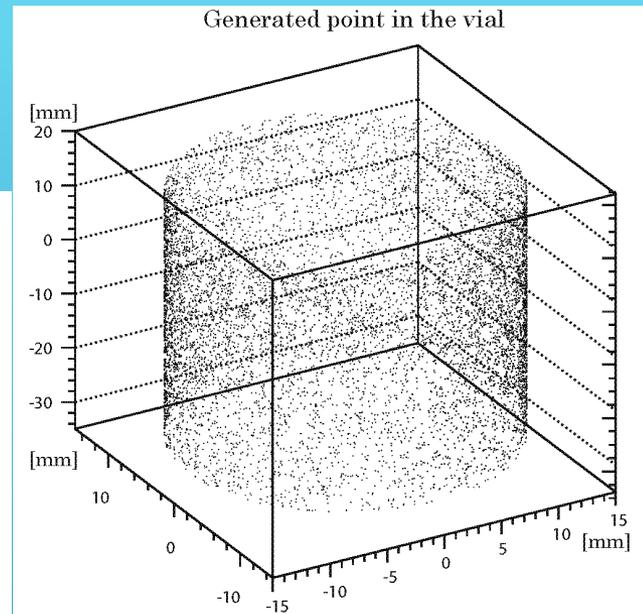
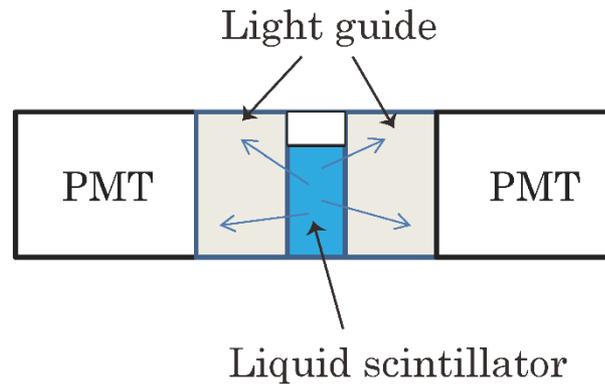
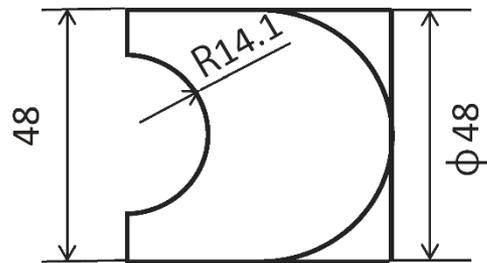
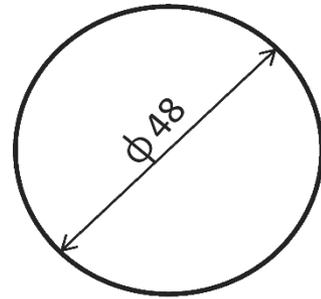
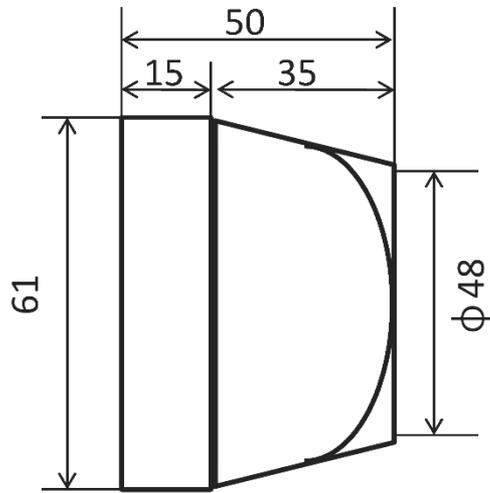
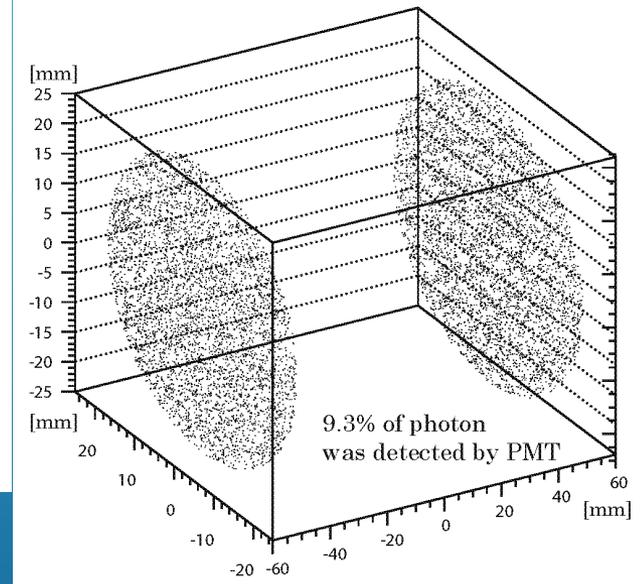


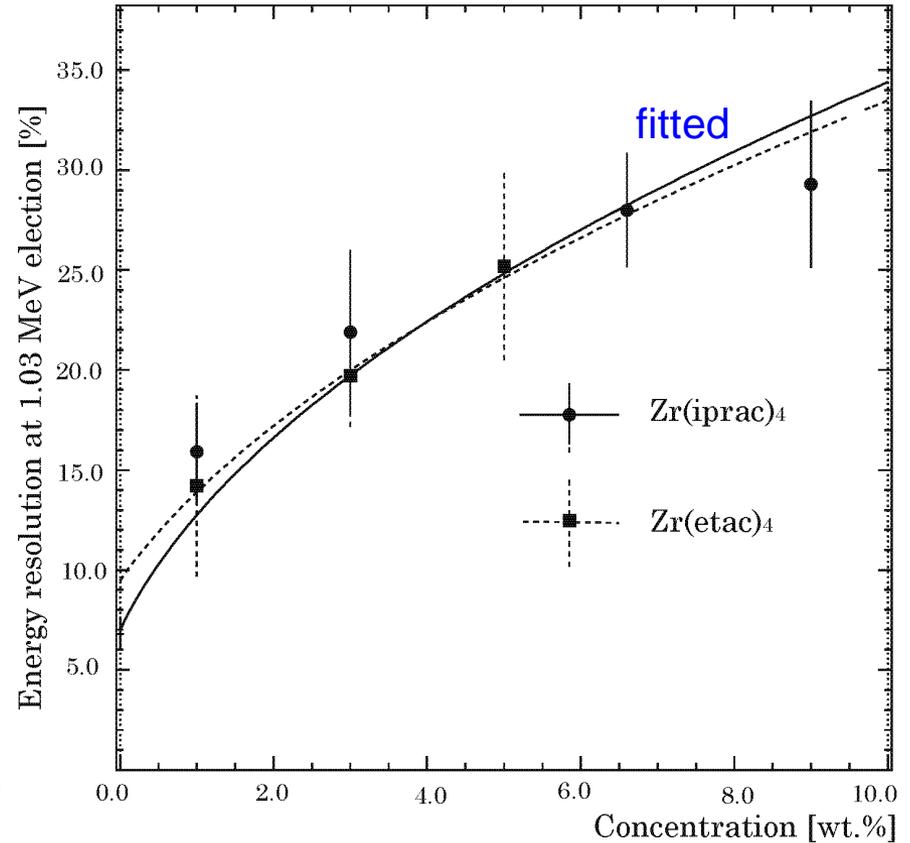
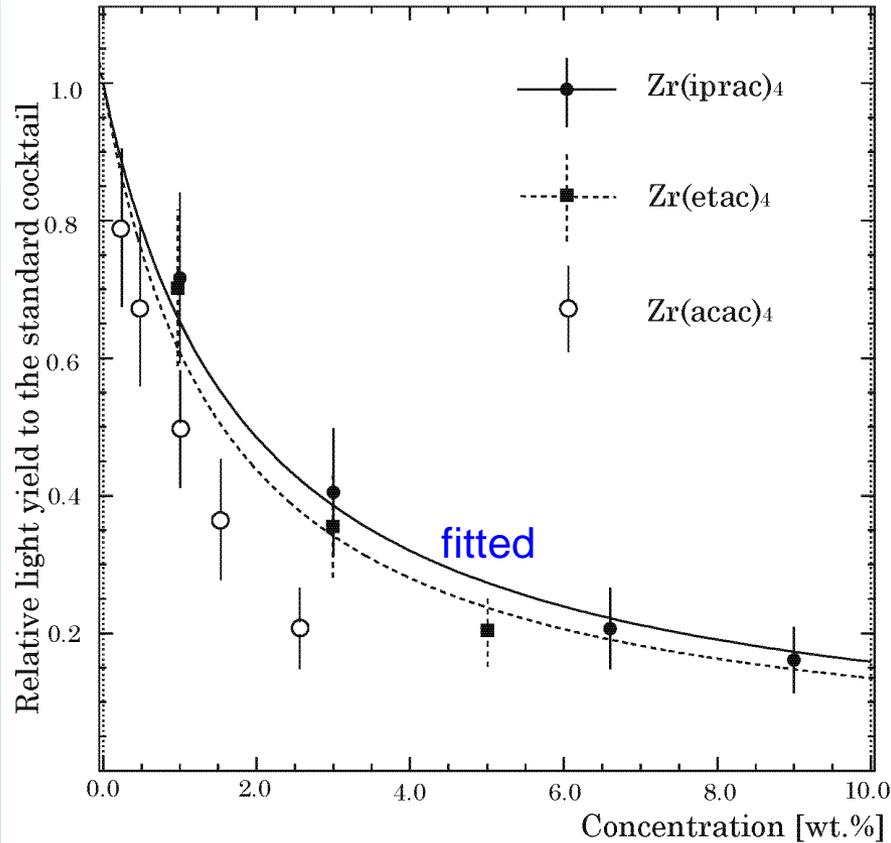
Photo coverage



detected scintillation light on PMT surface

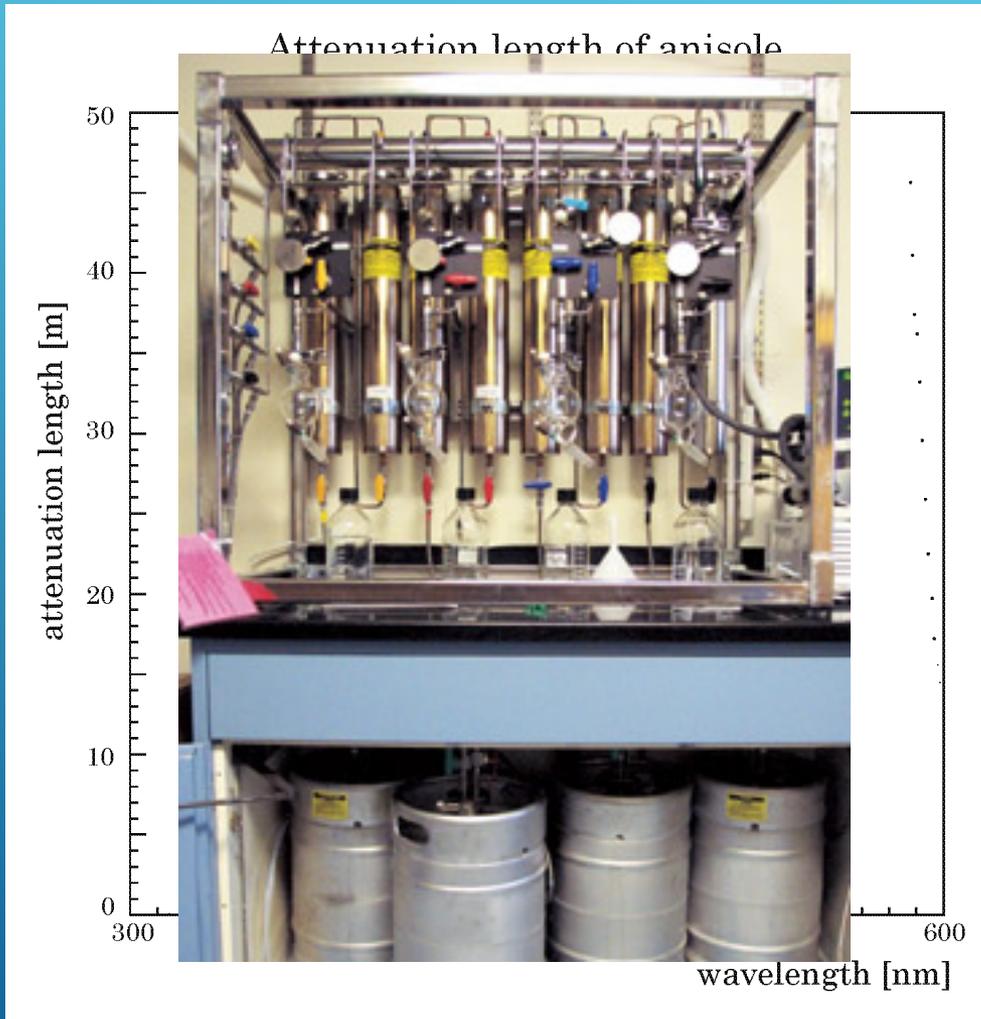


Light yield and energy resolution



Zr(ipcac)₄ and Zr(etac)₄ have almost same performance, but LY and Eres depend strongly on concentration.

PURIFICATION 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 Al_2O_3 . (Ref: [H.Grubbs et al., Org.Mat. 1996 15,1518-1520](#))