

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 XI

日本物理学会 2017年秋季大会

2017年9月13日

宮城教育大学教育学部 福田善之、龜井雄斗*、

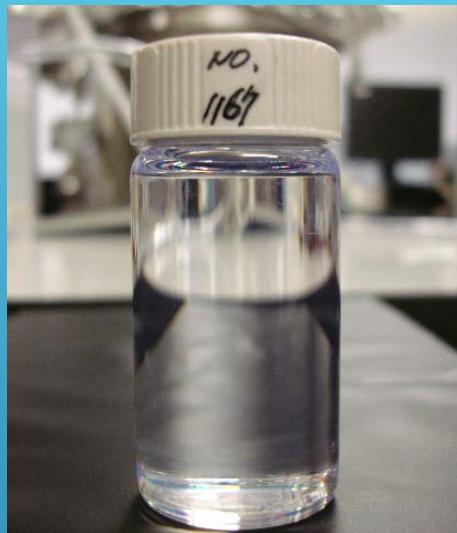
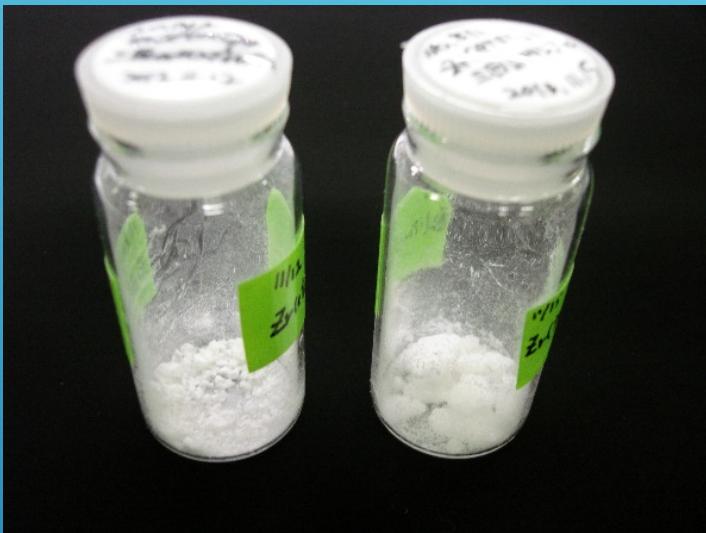
那仁格日樂*、小畠 旭*

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

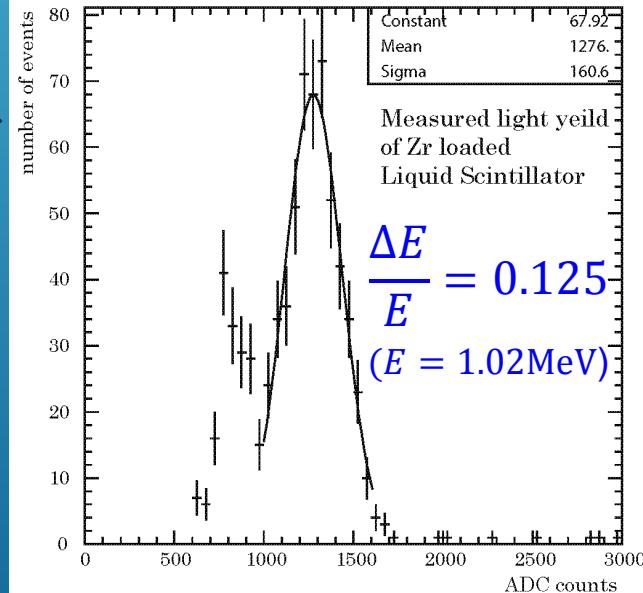
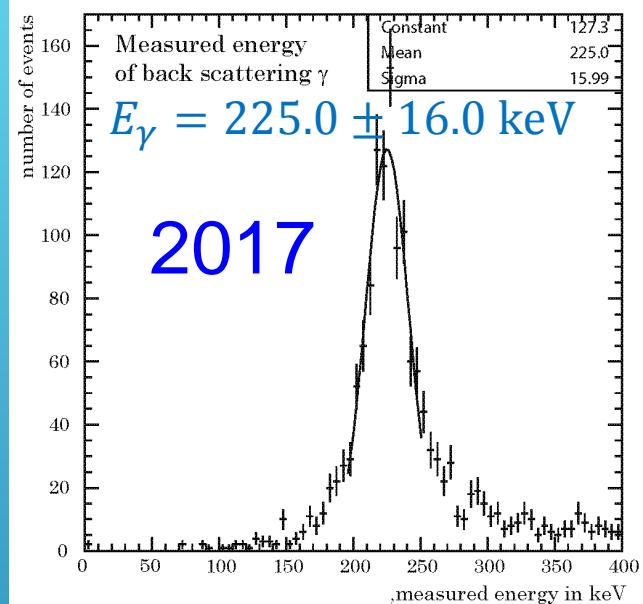
Zr loaded liquid scintillator



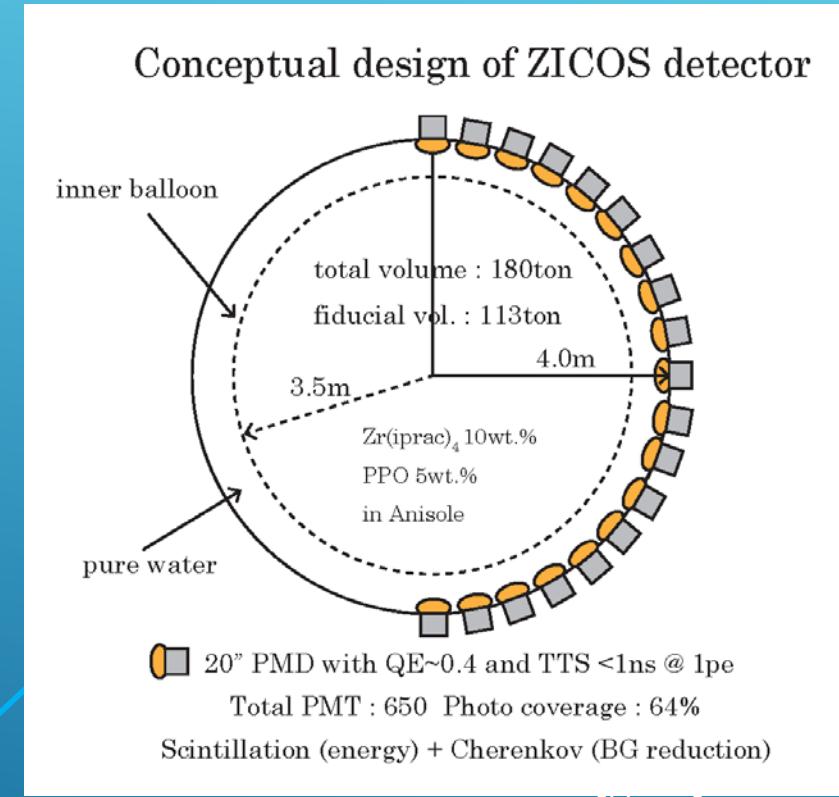
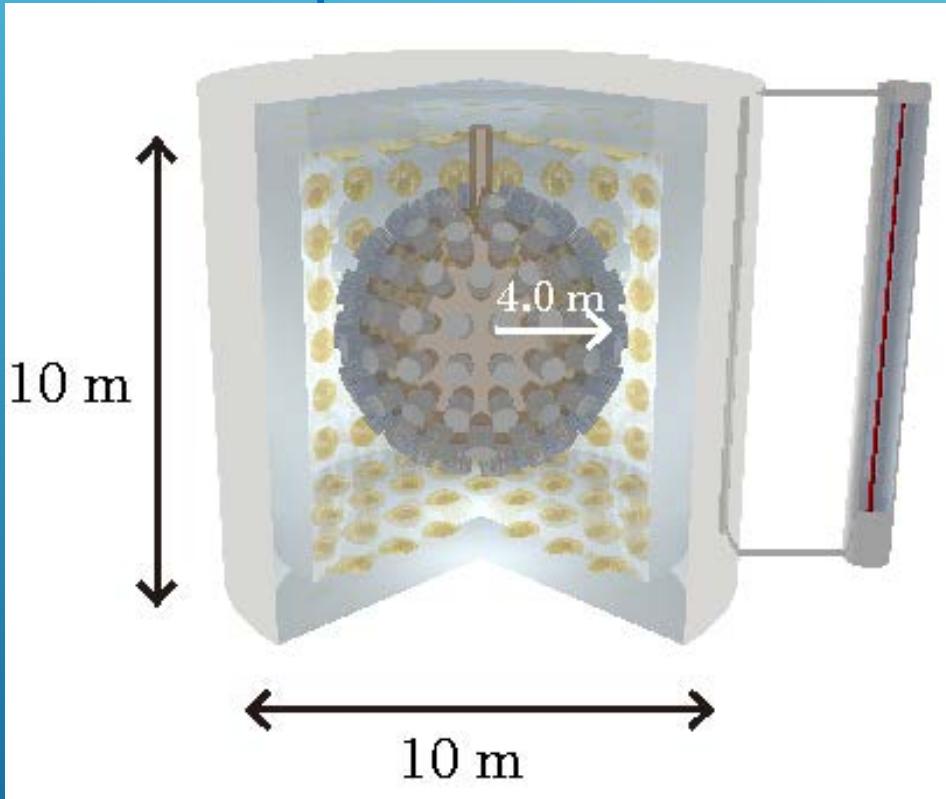
2013~2015

Measured on this August.

An energy resolution is obtained by $2.61 \pm 0.14\%$ at 3.35 MeV assuming 64% photo coverage of the photomultiplier.



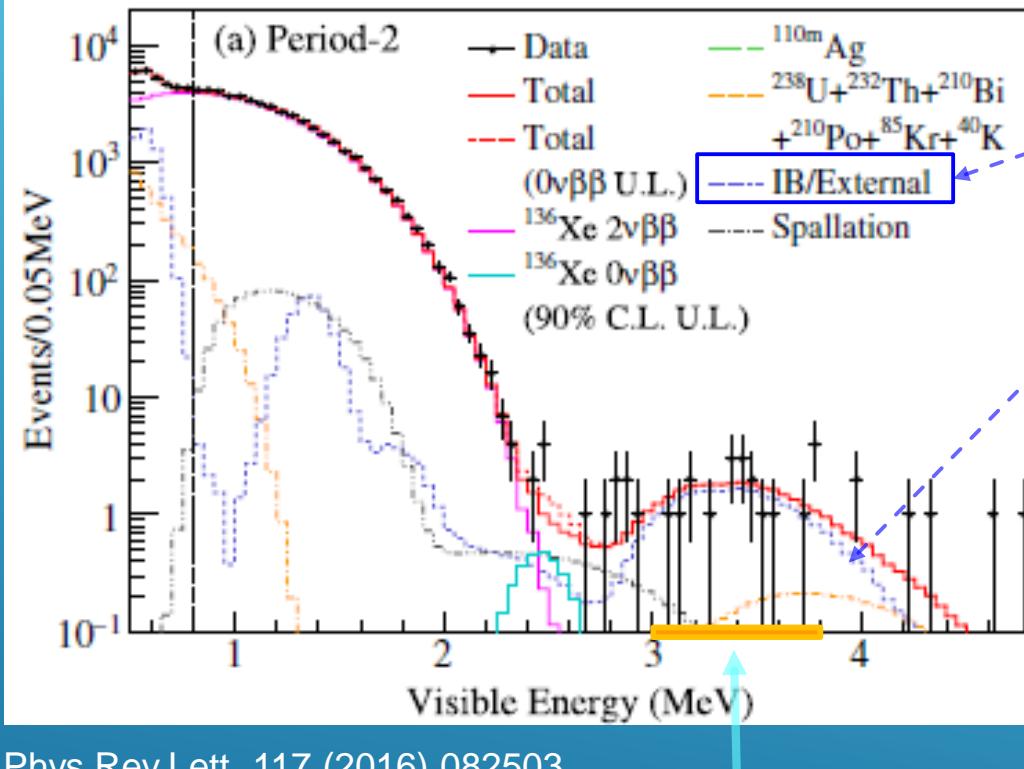
ZICOS- Zirconium Complex in Organic Liquid Scintillator for next generation of DBD experiment



$^{96}\text{Zr} \cdot 45\text{kg (not)} \rightarrow 865\text{kg (50\% enrich)}$
 $T_{1/2}^{0\nu}$ NEMO3 : $T_{1/2}^{0\nu} > 9.1 \times 10^{21} \text{ ys}$ ^6ys

Backgrounds around signal region

Measured by KamLAND-Zen



Phys.Rev.Lett. 117 (2016) 082503

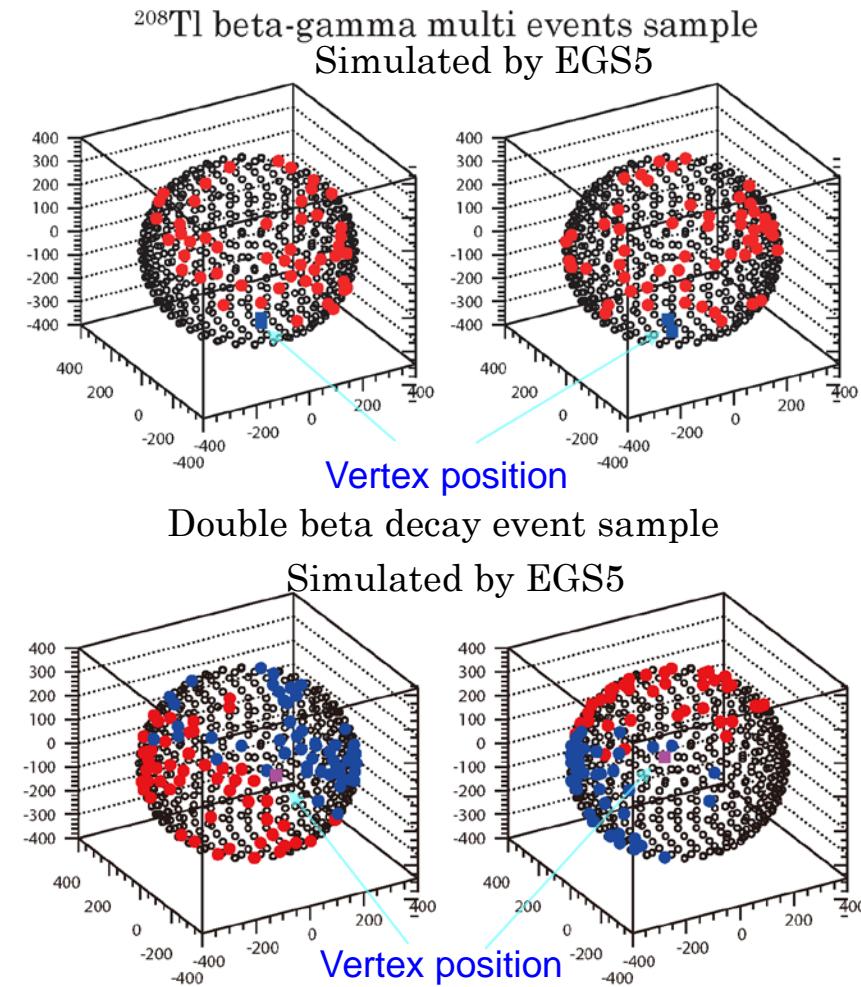
0νββ signal region for ^{96}Zr

^{208}TI on
surface of
balloon

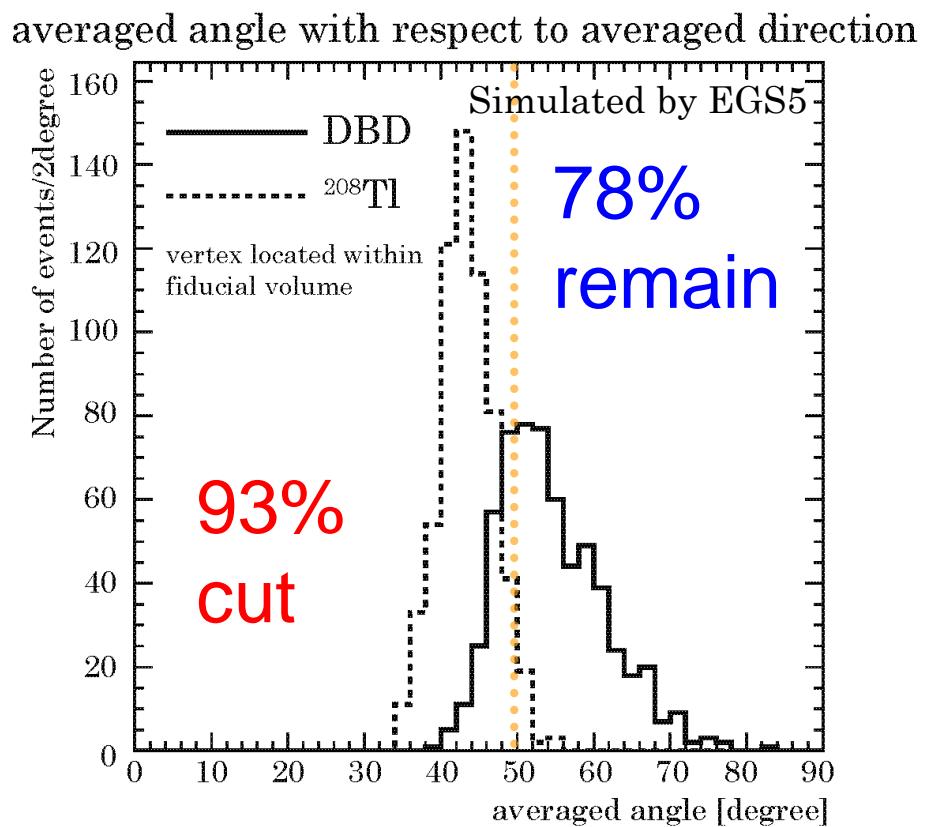
Even though using ultimate cleanliness like KamLAND-Zen, need further 1/20 ^{208}TI reduction for $T_{1/2}^{0\nu} > \sim 1 \times 10^{27} \text{ ys}$

An additional technique other than the energy spectral shape obtained by scintillation is necessary.

Hit pattern of Cherenkov lights

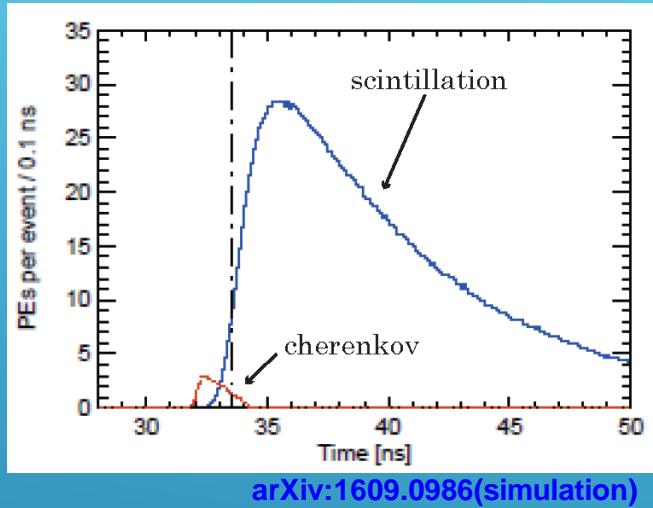


Hit pattern of Cherenkov lights for ^{208}Tl decay looks different from DBD.



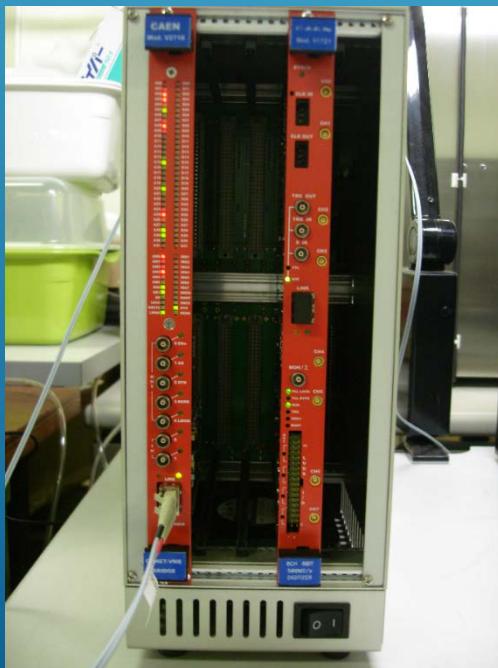
Averaged angle of ^{208}Tl decay is smaller values than that of DBD.

Separation of Cherenkov and Scintillation



Difference of light emission mechanism

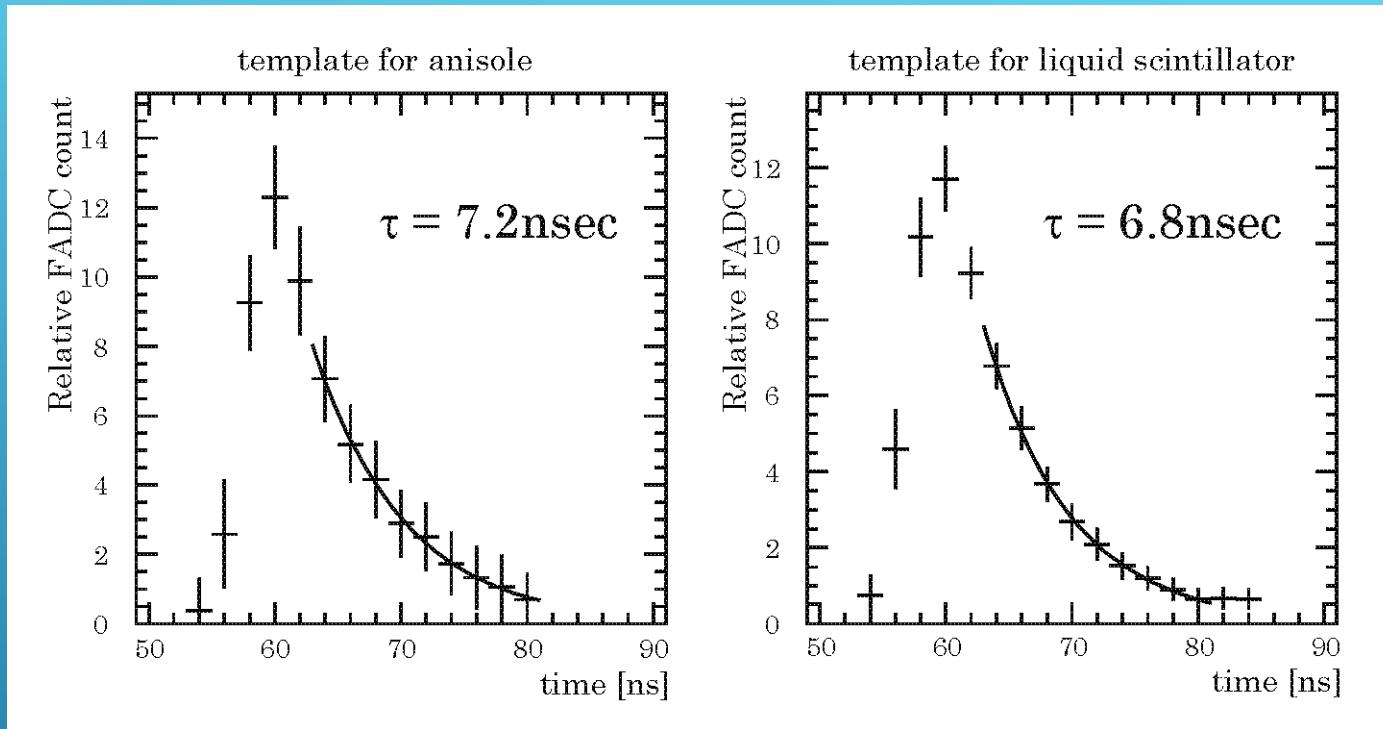
- Cherenkov : an order of a few 100 pico second due to the electro-magnetic process
- Scintillation : an order of tenth nano seconds in general.



Possible to extract PMT hits received Cherenkov lights by Pulse Shape Discrimination.

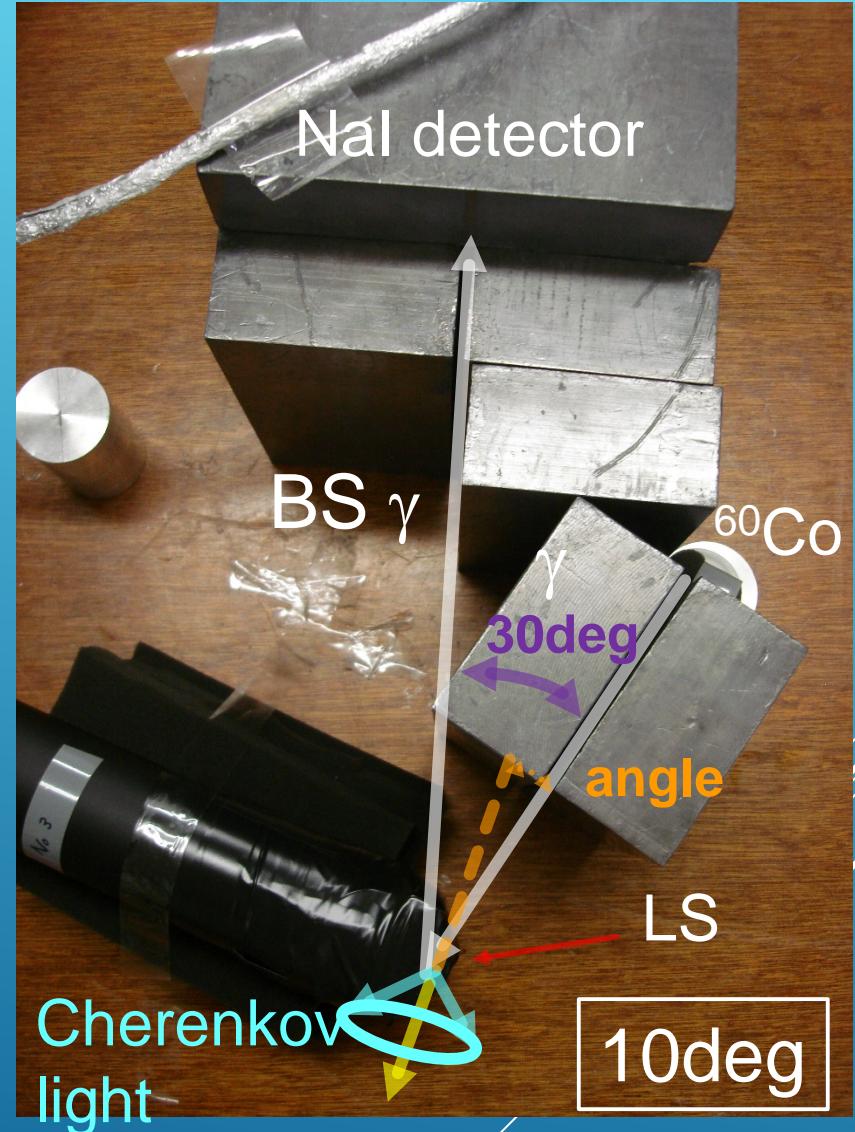
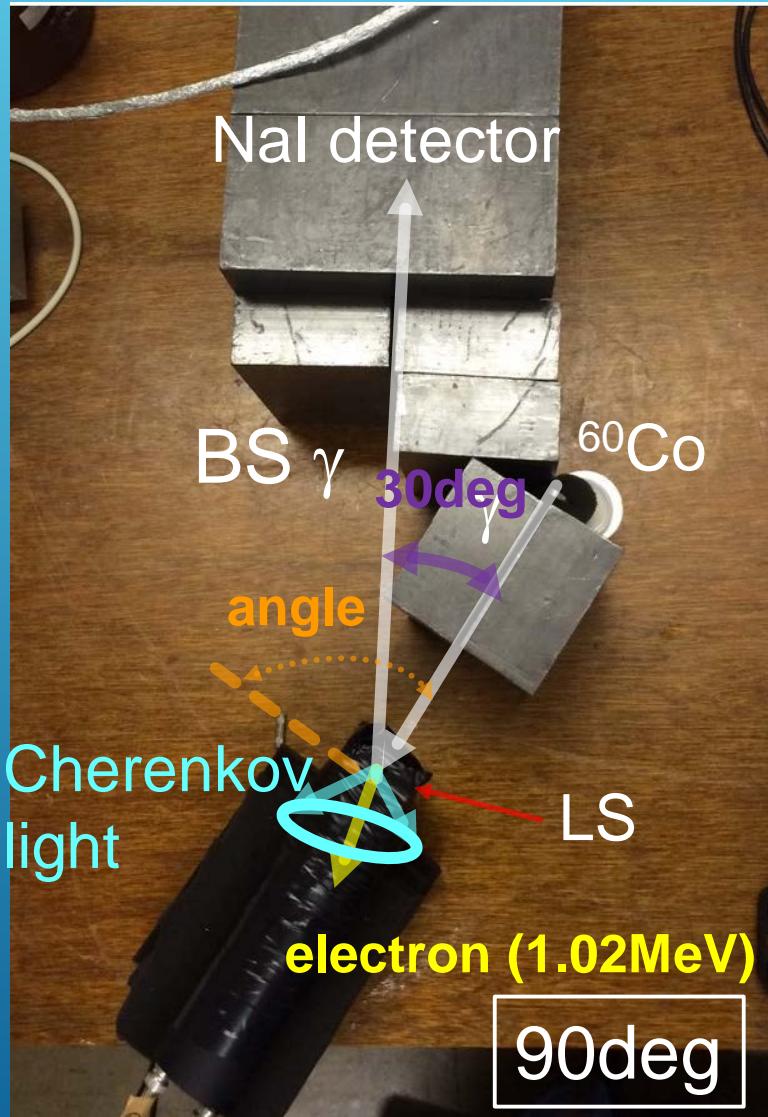
- CAEN V1721 8 channel 8bit 500MS/s FADC digitizer
- CAEN V2718 VME-PCI Optical Link Bridge

Pulse shape of scintillation light

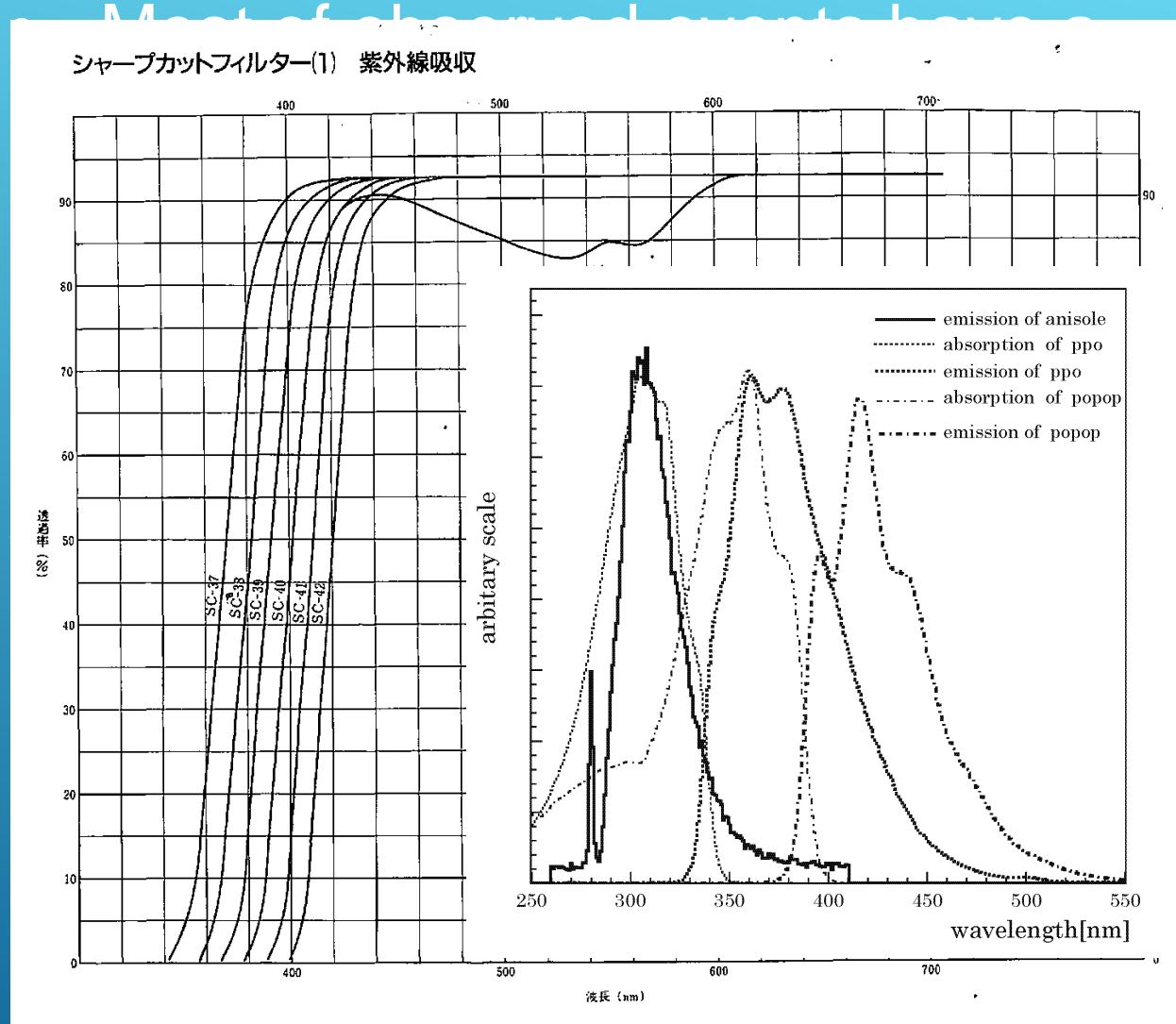
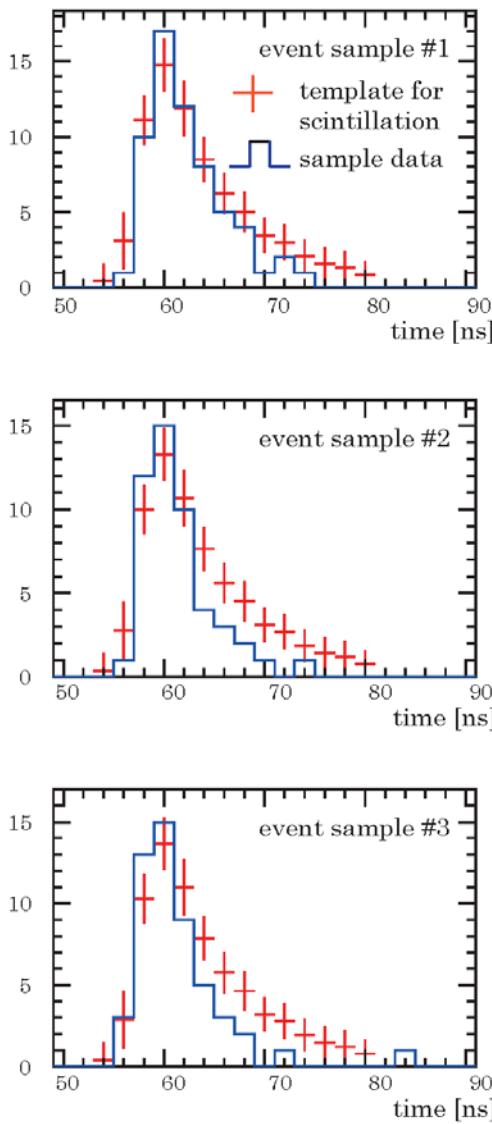


- Templates of FADC timing pulse shape for **scintillation light** were obtained for both anisole and Zr loaded liquid scintillator.
- Both decay times are almost same, and it was about 7ns.

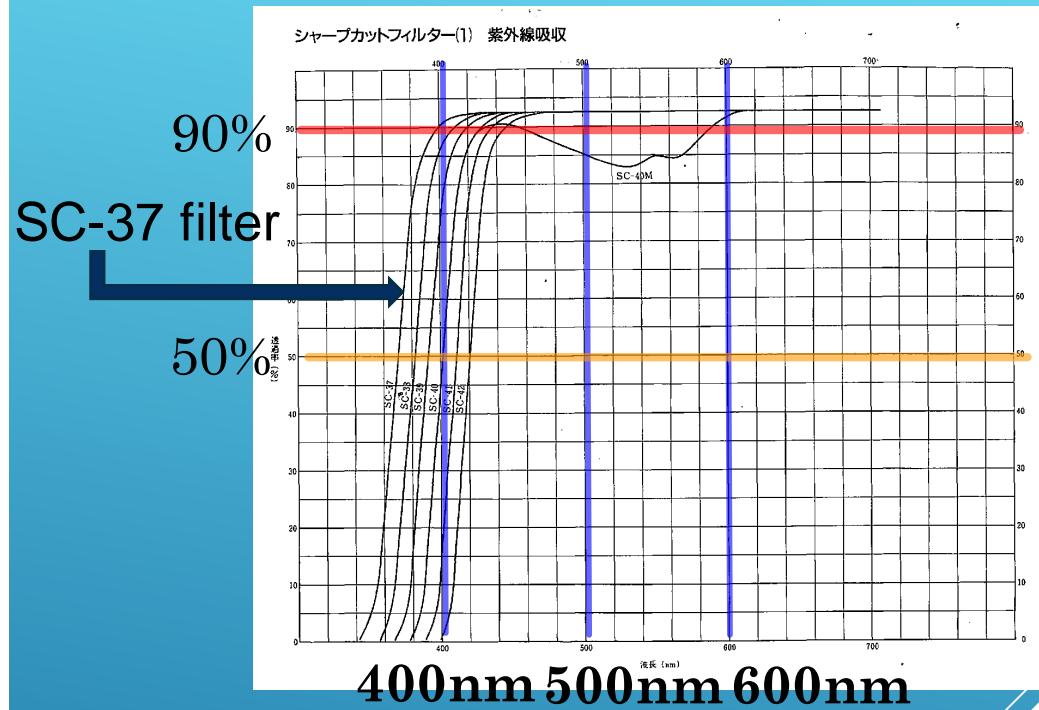
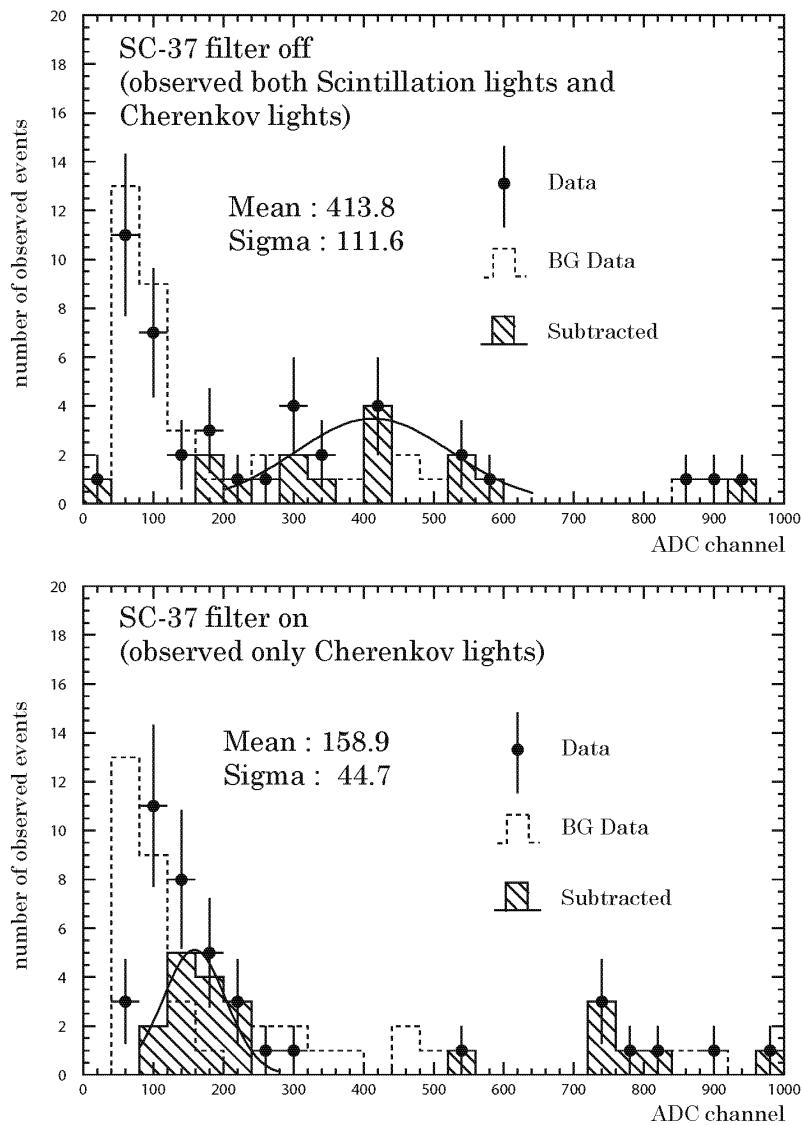
Observation of Cherenkov lights



Pulse shape observed in Anisole

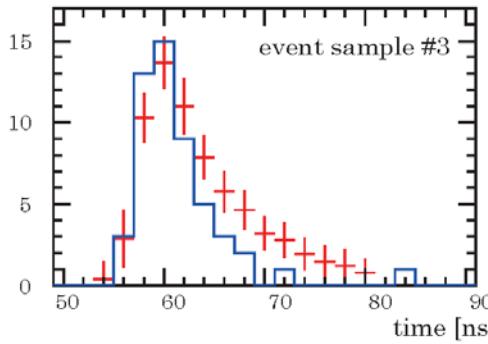
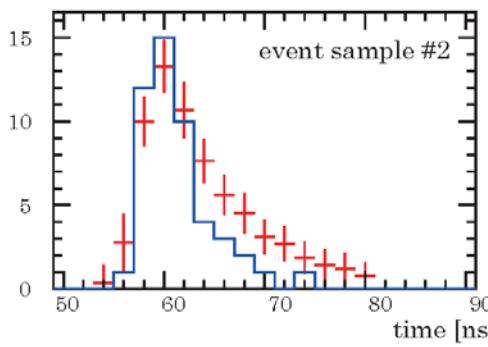
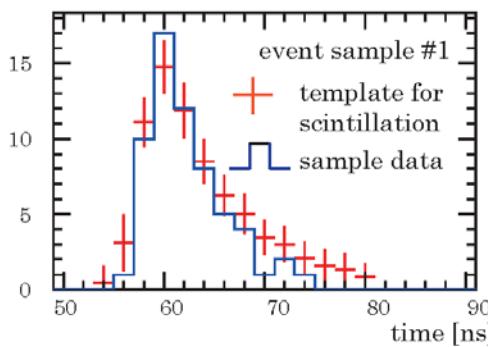


Light yield of Cherenkov lights



Cherenkov light yield ($\lambda > 400\text{nm}$)
Scintillation light yield of anisole
 $= \sim 0.02 \equiv \sim 200 \text{ photon/MeV}$

Pulse shape observed in Anisole

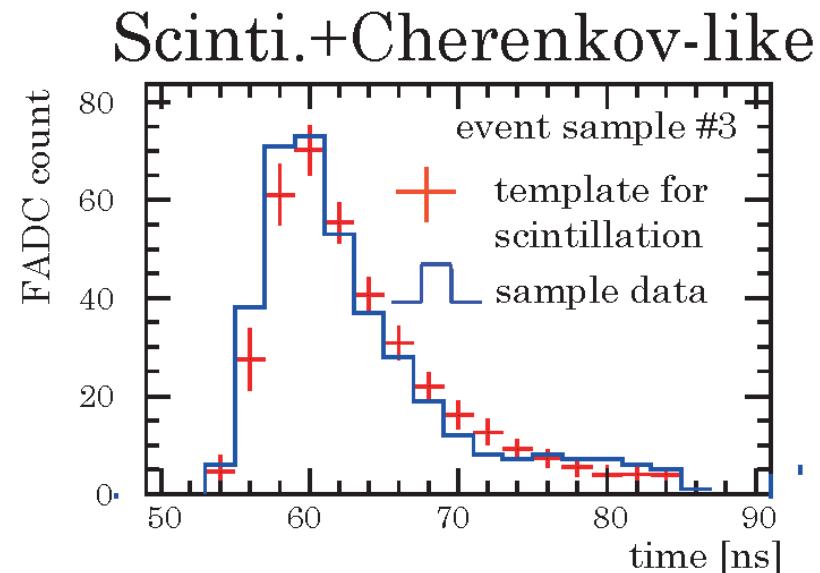
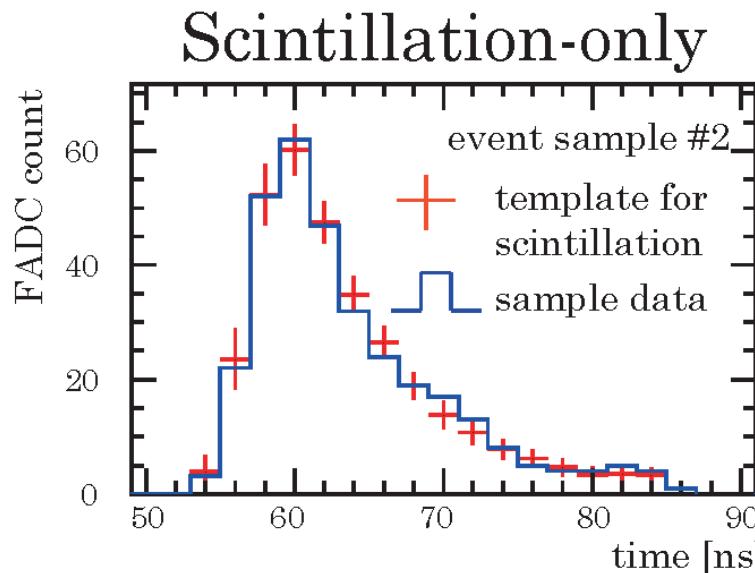


- Most of observed events have a different shape from scintillation.
- Same shape was observed in pure water.
- Recently same shape was found even though using **UV cut filter**.
- Observed directionality : 6 events (90deg) and 1 events (10deg)



These pulse shapes might consist of **Cherenkov lights**.

Pulse shape observed in Zr loaded LS



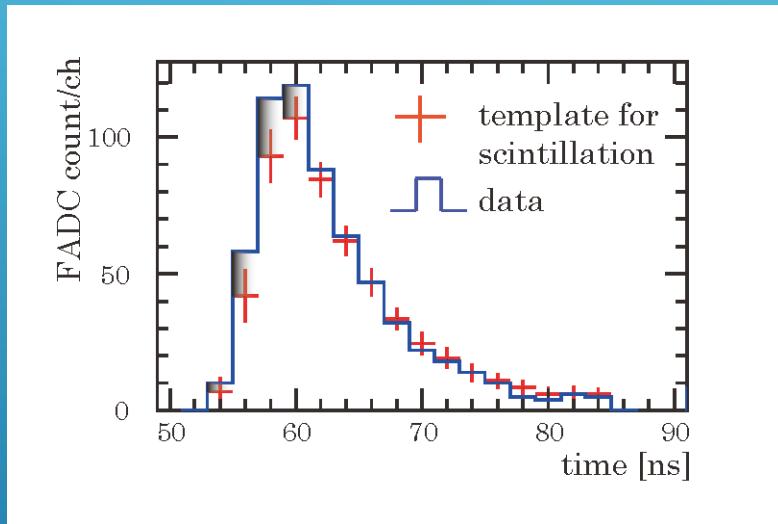
Mainly two types of shape (Scintillation-only and Scintillation + Cherenkov-like) were observed.



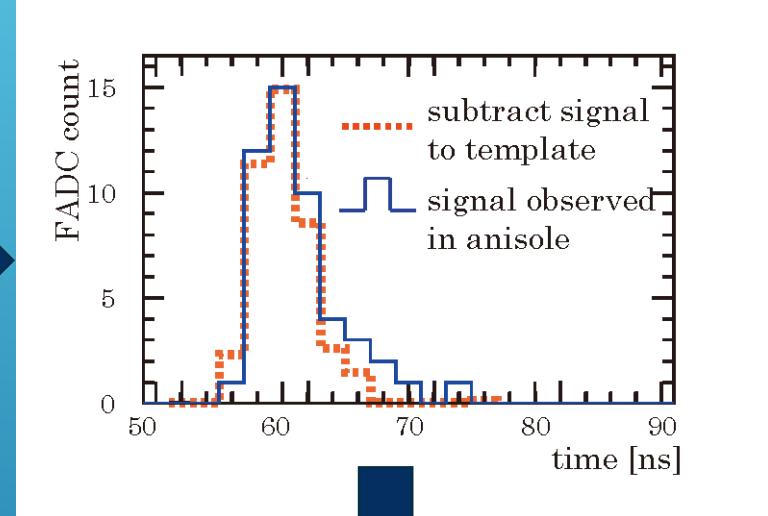
Is this excess caused by Cherenkov light?

Excess of rise time shape

Typical shape of Scinti.
+ Cherenkov-like



Excess shape and shape
observed in anisole

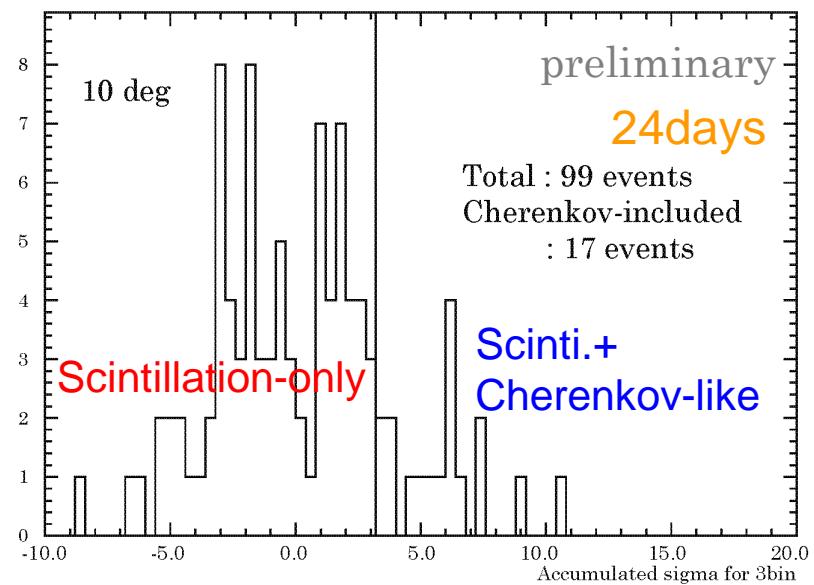
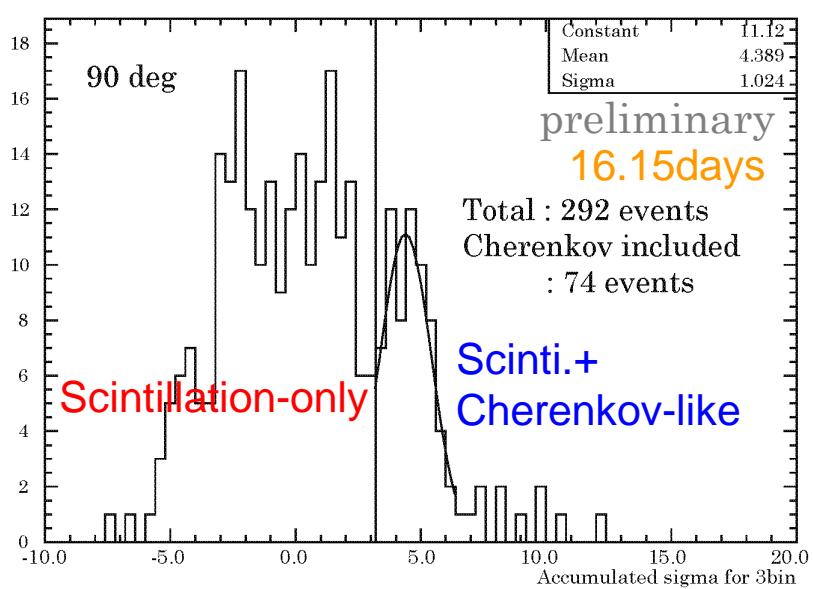


Excess shape is similar to the one observed in anisole.

It might be caused by Cherenkov lights!

Pulse shape discrimination whether the event includes
Cherenkov-like or not using excess could be possible.

Pulse shape discrimination



Count rate for events included Cherenkov-like

- 90deg 4.6 events/day
- 10deg 0.7 events/day

Significance : 6.0 σ

Same directionality as observed in anisole.

However, the number of events for scintillation-only also decreased at 10 degree.



Need more data and detailed analysis to confirm above results.

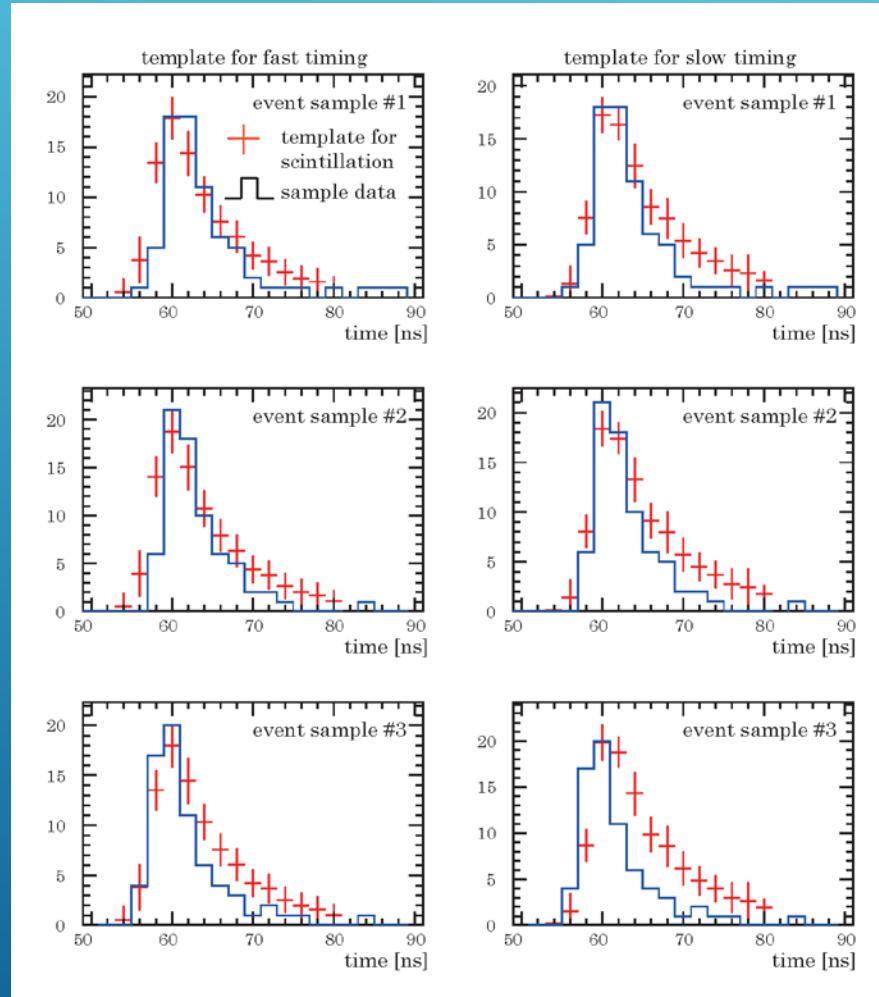
Summary

- ▶ Conceptual design of ZICOS detector (10 wt.% Zr(iprac)₄ loaded Liquid Scintillator has 2.6% @3.35MeV energy resolution assuming 64% photo coverage of 20" PMT) for next generation DBD experiment. ($T_{1/2}^{0\nu} > \sim 10^{27}$ years).
 - ^{96}Zr : 45kg (nat.) → 865kg(50% enrich)
 - Further 1/20 reduction of ^{208}Tl backgrounds using PMT hit pattern of Cherenkov lights.
- ▶ Cherenkov might have a different pulse shape from scintillation even though O(1MeV) electron.
- ▶ PSD could be useful for the extraction of Cherenkov lights, however still need to study.

BACKUP

Pulse shape observed in H_2O

90deg



- Same pulse shape of timing as anisole was observed.
- This signal should be caused by Cherenkov light, because of no scintillation in H_2O .

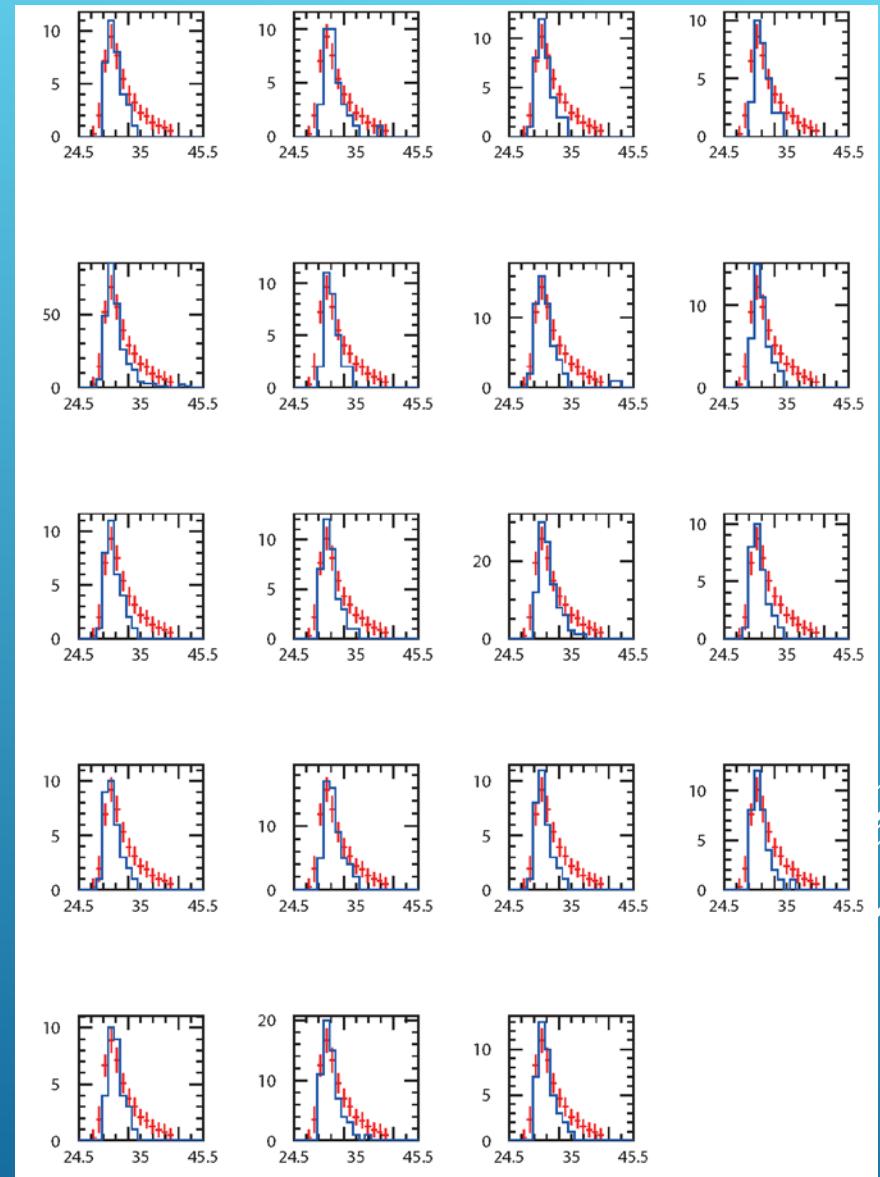


This pulse shape is made by Cherenkov lights.

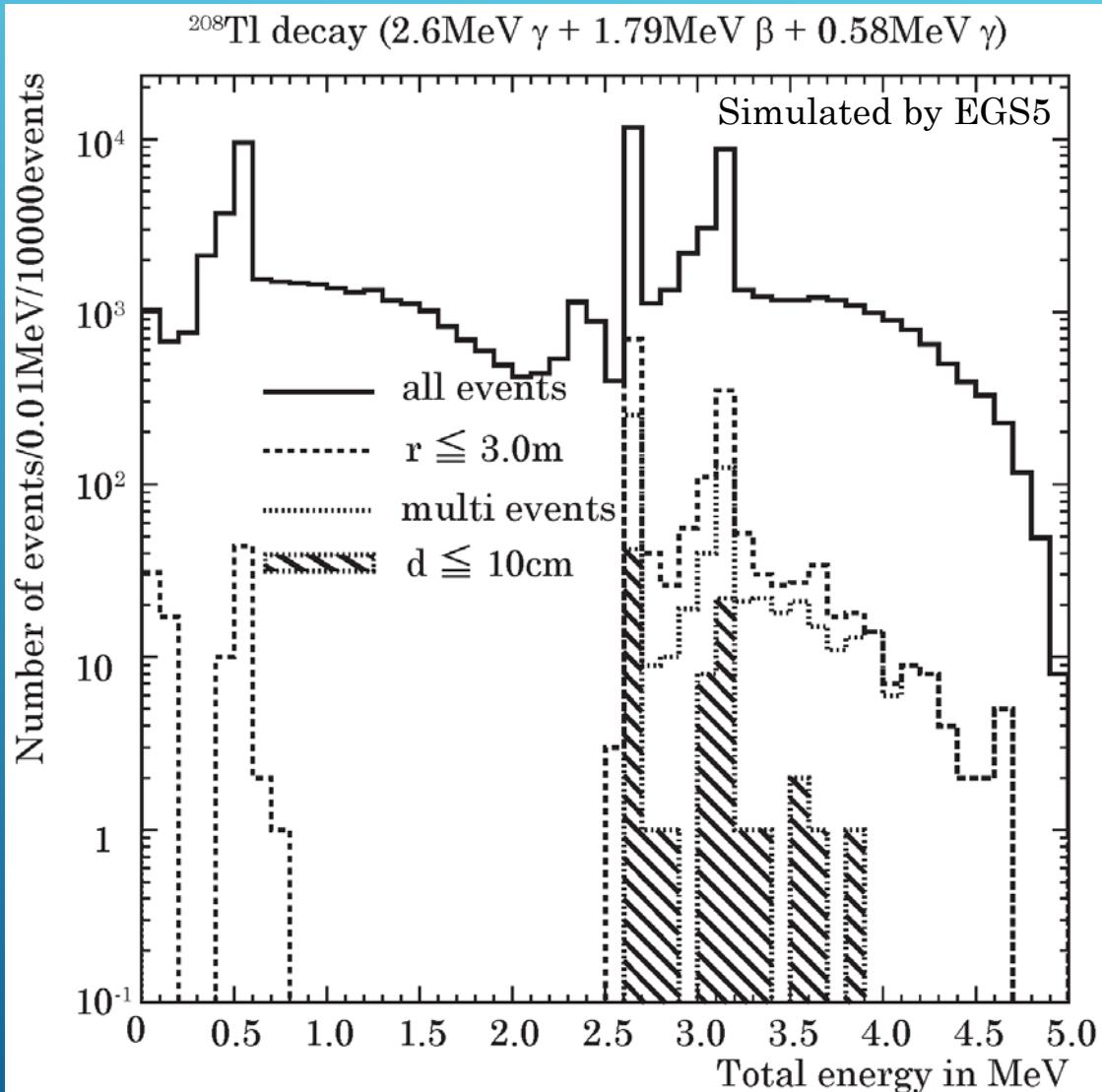
Pulse shape observed in anisole with UV cut filter SC-37

Observed events except large pulse height (only 2 events) has same pulse shape (or much narrow) as before.

Scintillation should not be measured due to UV cut filter, so confirmed that this shape consists of Cherenkov light.



Reduction of ^{208}Tl decay

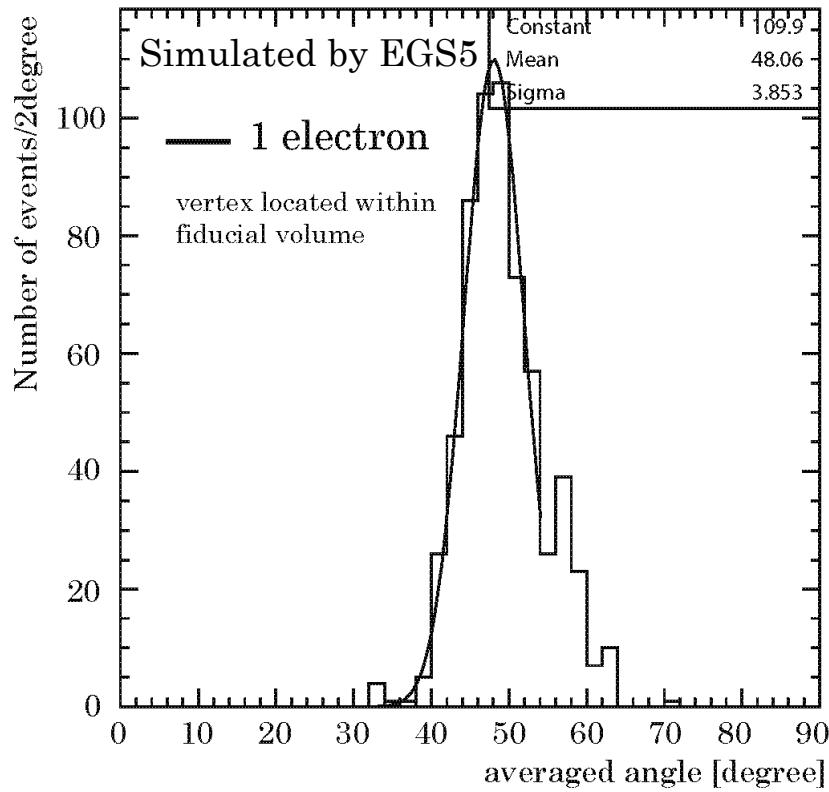


- 1) $E : 3.0\text{-}3.7\text{MeV}$
17925 events
- 2) Fiducial volume
628 events
- 3) Multi events
263 events
- 4) Closer events
($d \leq 10\text{cm}$)
35 events

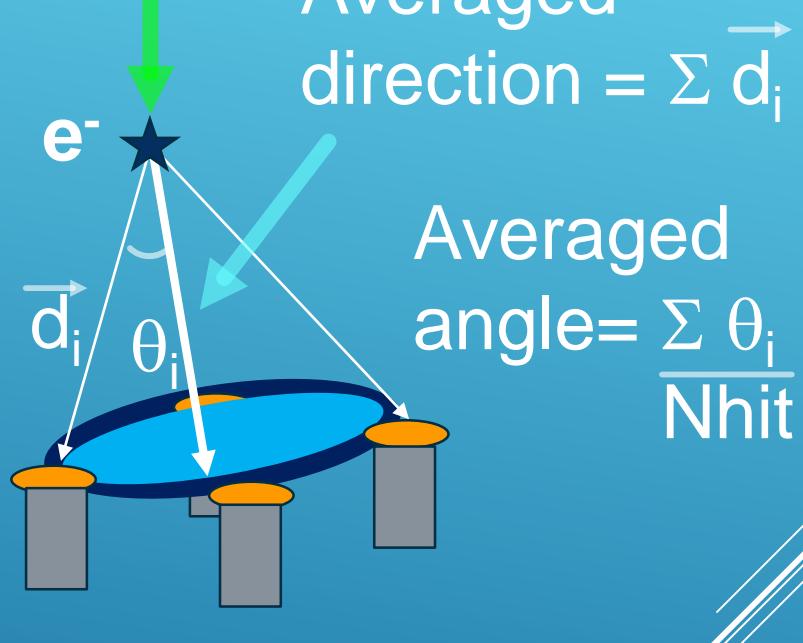
~1/20 BG reduction
could be achieved by
using the information
from Cherenkov light.

Averaged angle of Cherenkov hit

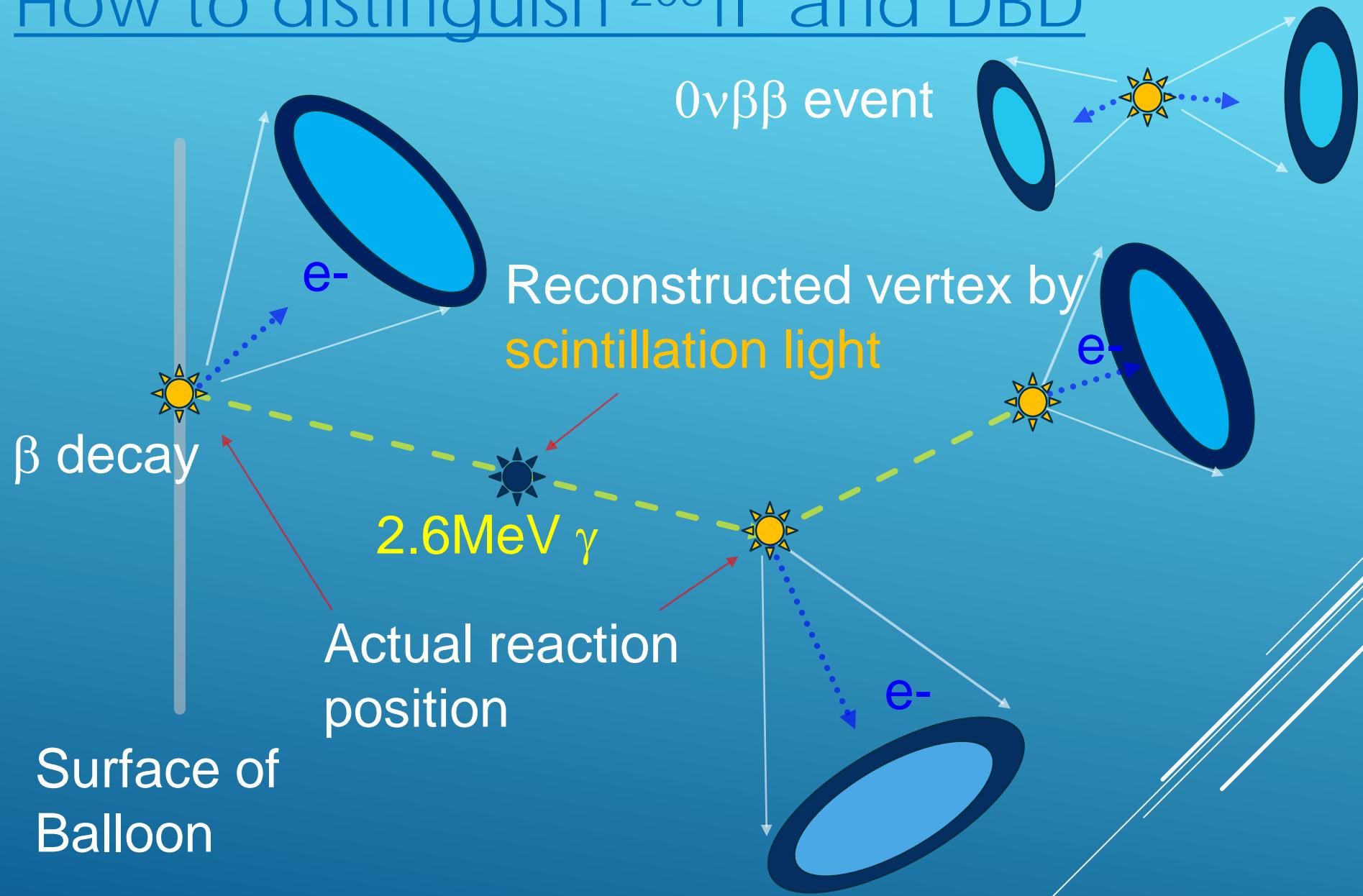
averaged angle with respect to averaged direction



Vertex position obtained by scintillation



How to distinguish ${}^{208}\text{TI}$ and DBD



Neutrino mass sensitivity of ZICOS experiment

Total mass : 180ton (fiducial volume : 113ton)

Measurement time: 2 years

10wt.% Zr(iprac)₄ = 12.6ton includes 1.7ton of
Zirconium = 45 kg of ⁹⁶Zr (**natural abundance 2.6%**)

$T_{1/2}^{0\nu} > 4 \times 10^{25} \text{y}$ ← Not enough for $0\nu\beta\beta$ search

1) Zr enrichment

50% enrichment of ^{96}Zr (e.g. 57.3% for NEMO-3)

^{96}Zr will be 865kg then $T_{1/2}^{0\nu} > \sim 2 \times 10^{26}\text{y}$

2) BG (^{208}TI) reduction

BG level < 1/20 × KL-Zen

then $T_{1/2}^{0\nu} > \sim 1 \times 10^{27} y$ Today's talk

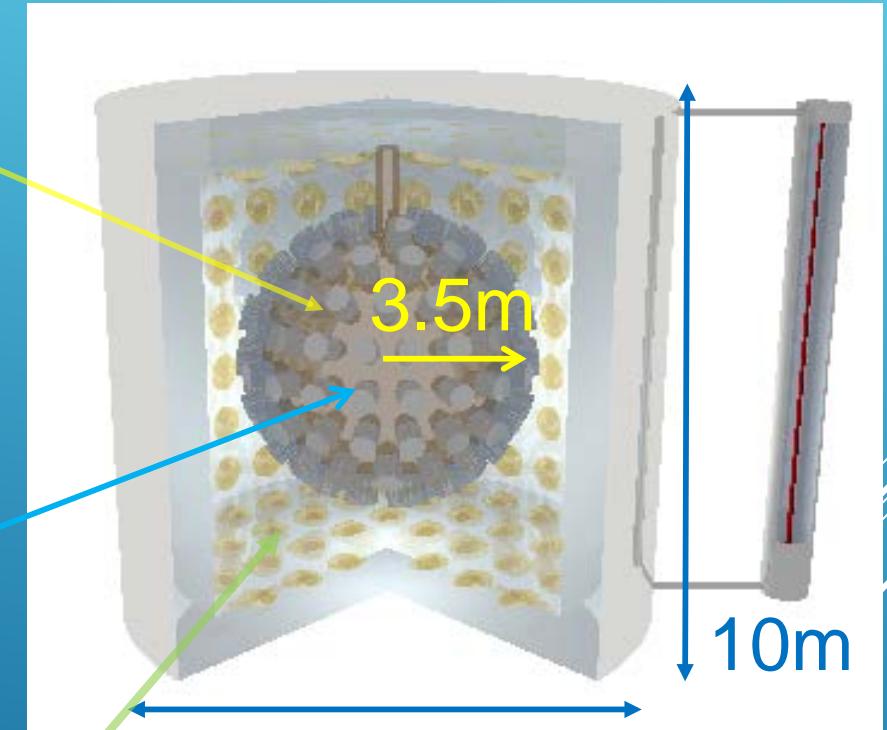
ZICOS- Zirconium Complex in Organic Liquid Scintillator for neutrinoless double beta decay

1.5wt.% Zr loaded Liquid Scintillator :

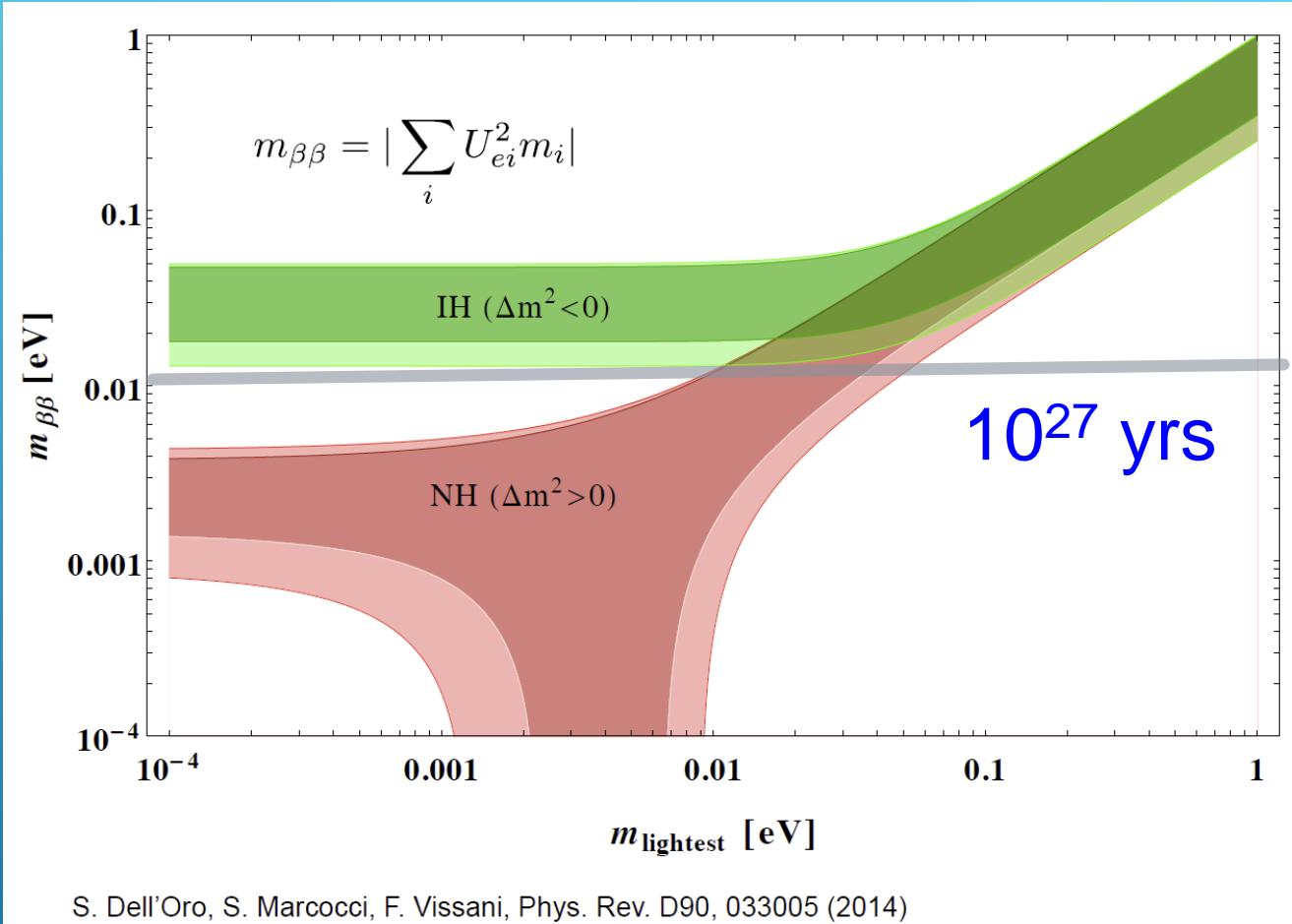
Light yield of $48.7 \pm 7.1\%$ for BC505,
and an energy resolution of $2.8 \pm 0.4\%$
at 3.35 MeV assuming 64% photo coverage of the photomultiplier

Inner detector : 64% photo coverage with 20" ultra-high spec. PMT including 1.7ton Zirconium loaded 113 tons LS in fiducial volume. (Total vol. : 180 tons)

Outer detector : active veto using pure water surrounding inner detector in order to veto muons and external γ -ray backgrounds.



Future $0\nu\beta\beta$ experiments

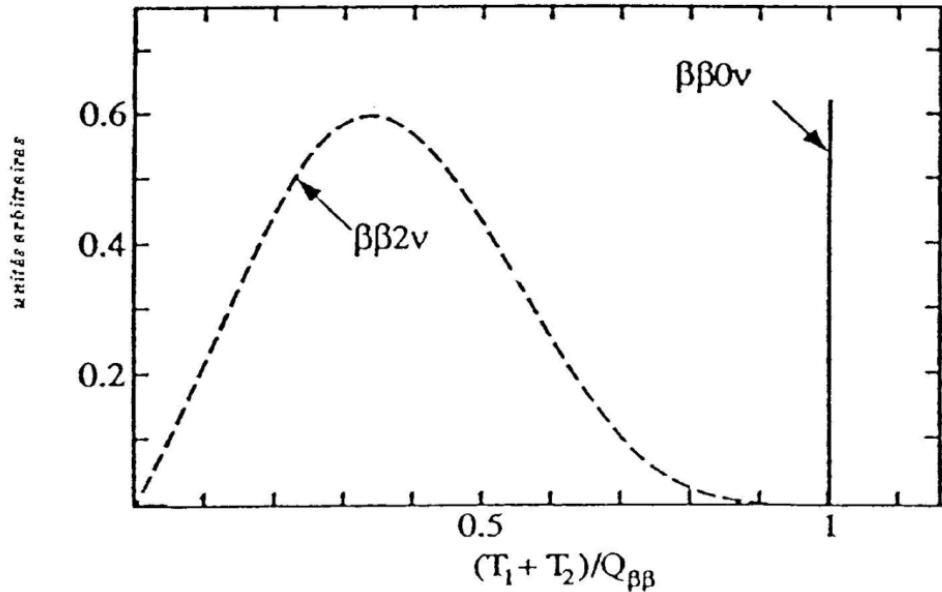


~tons of target and ~zero BG detector will be necessary for next generation $0\nu\beta\beta$ experiment.

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



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

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

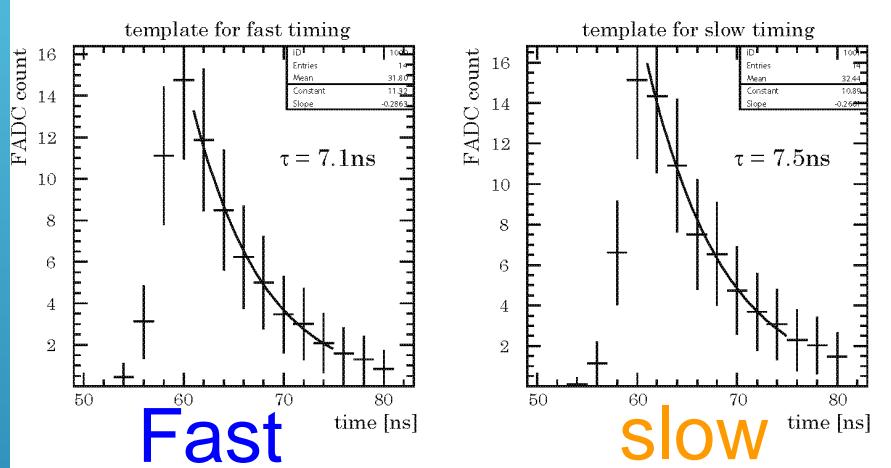
a: abundance M: target mass

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

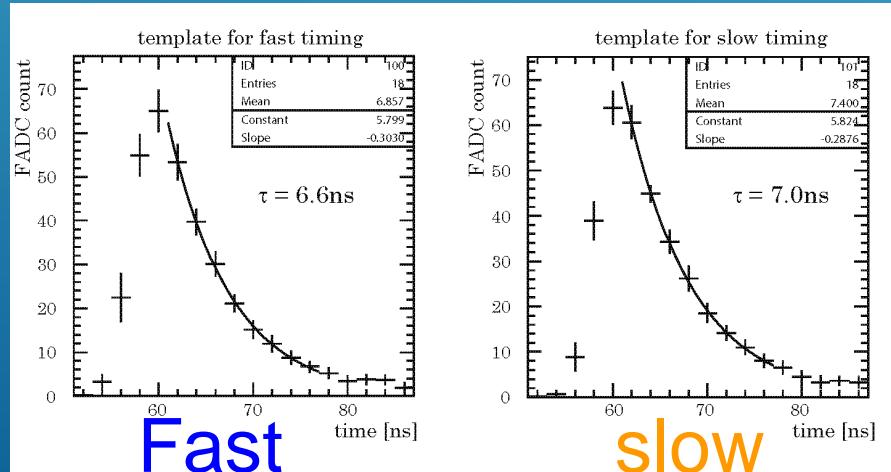
Requirement : Low BG, Large target mass, High E-resolution

Pulse shape of timing information

Anisole only



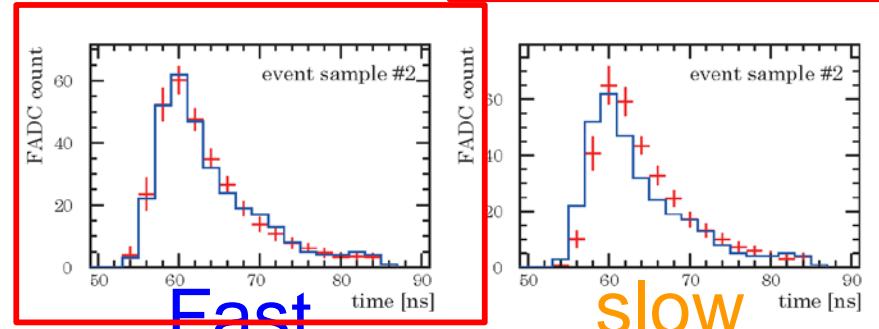
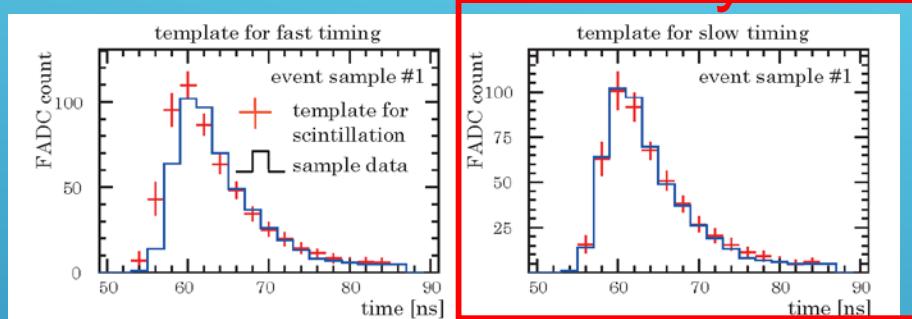
Zr(ipr₃)₄ loaded LS



- Templates of pulse shape of timing information for scintillation light were obtained by FADC.
- Fast and slow rise time component were observed due to FADC resolution.
- Both decay time of scintillation light are about 7ns.

Pulse shape observed in Zr loaded LS

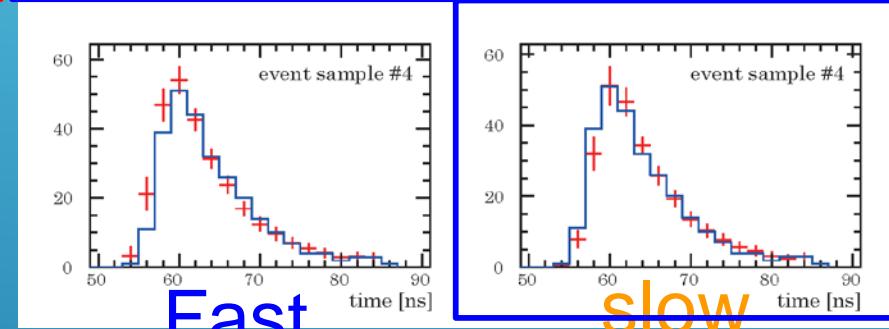
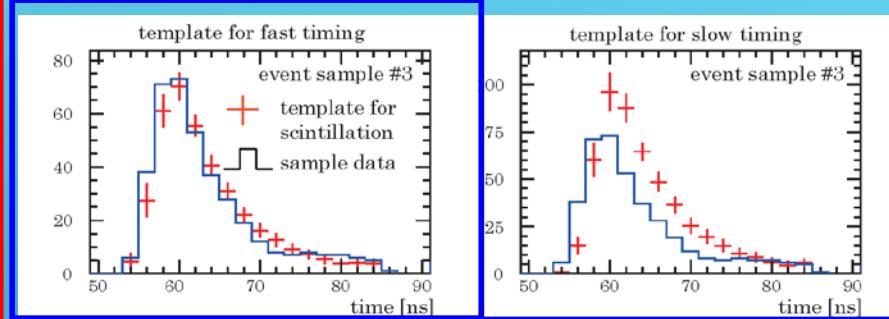
Scintillation-only



Fast

slow

Cherenkov-included



Fast

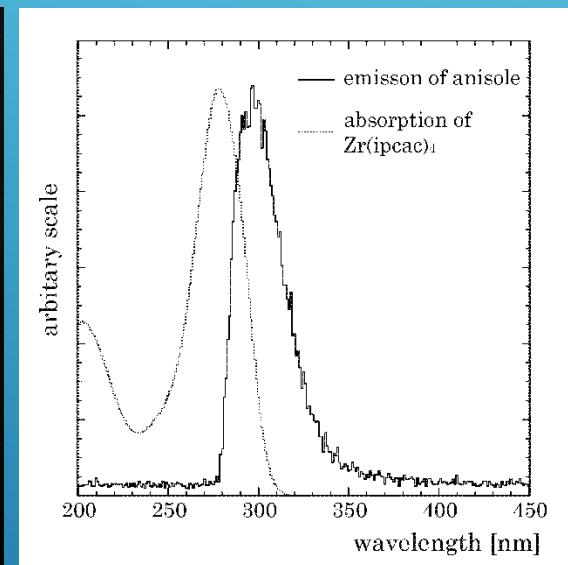
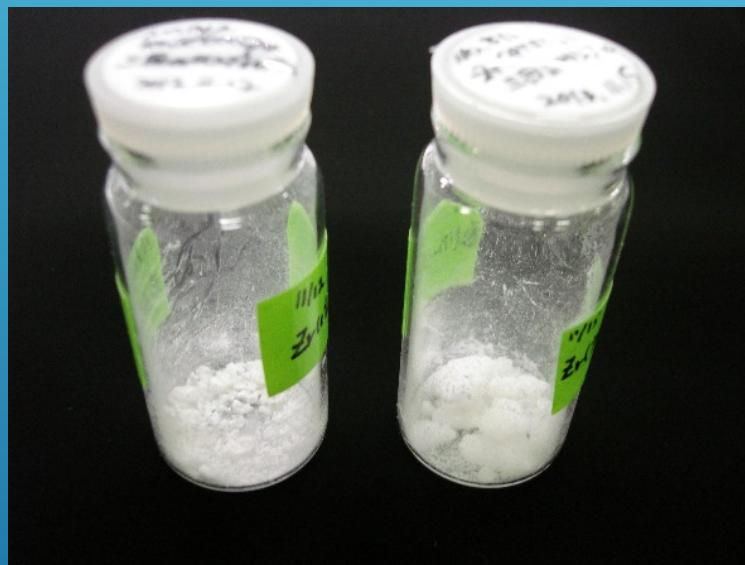
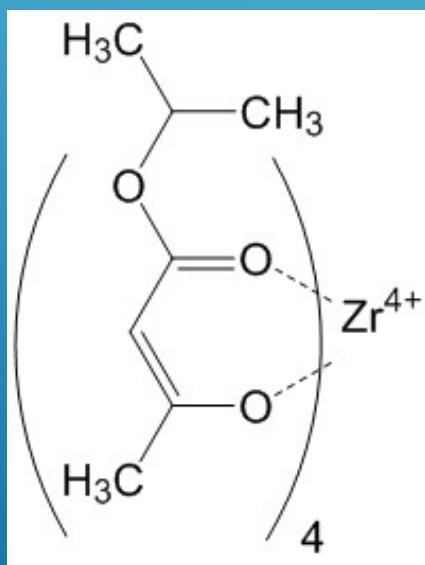
slow

Two types of pulse shape (Scintillation-only and Cherenkov-included) were observed.

We can use pulse shape discrimination for selection of events which include Cherenkov lights.

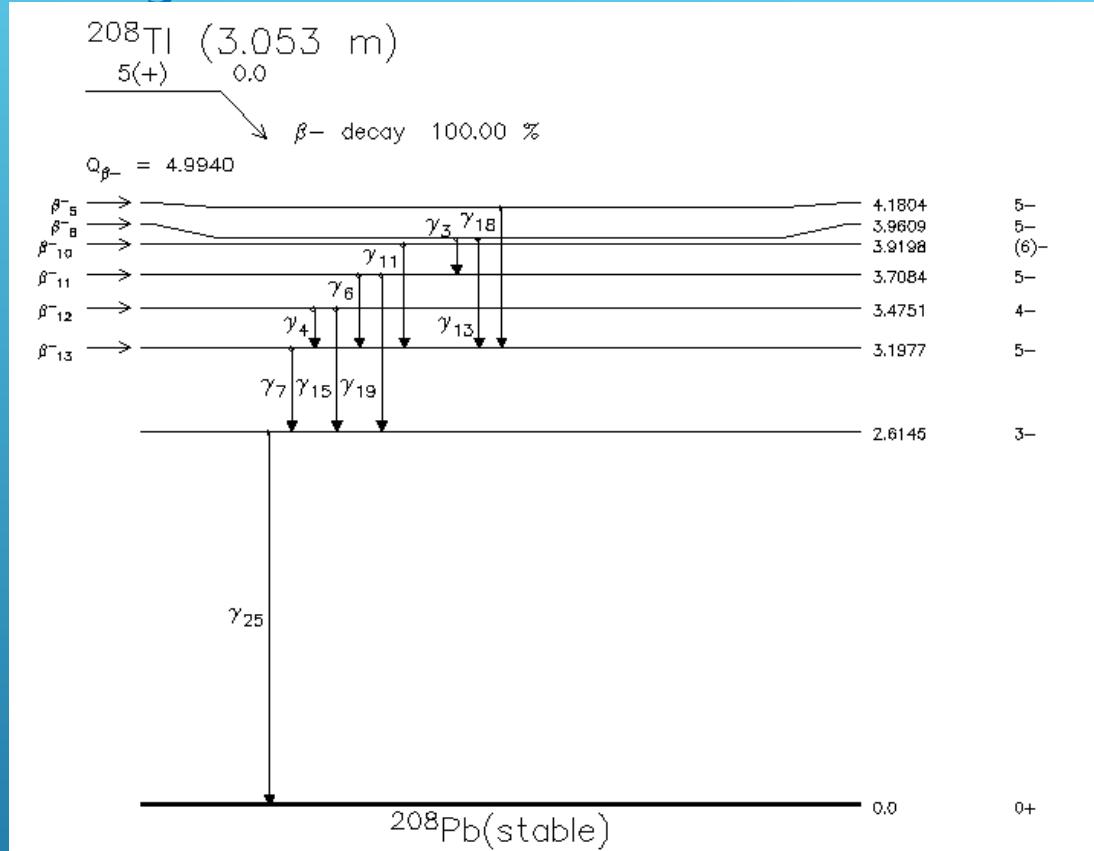
tetrakis(isopropyl acetoacetate) zirconium

$\text{Zr}(\text{CH}_3\text{COCHCOOCH(CH}_3)_2)_4 : \text{Zr(iprac)}_4$
Molecular weights : 663.87



LS = Zr(iprac)_4 : 10 wt.% PPO: 5wt.%
(POPOP: 0.05wt.%)

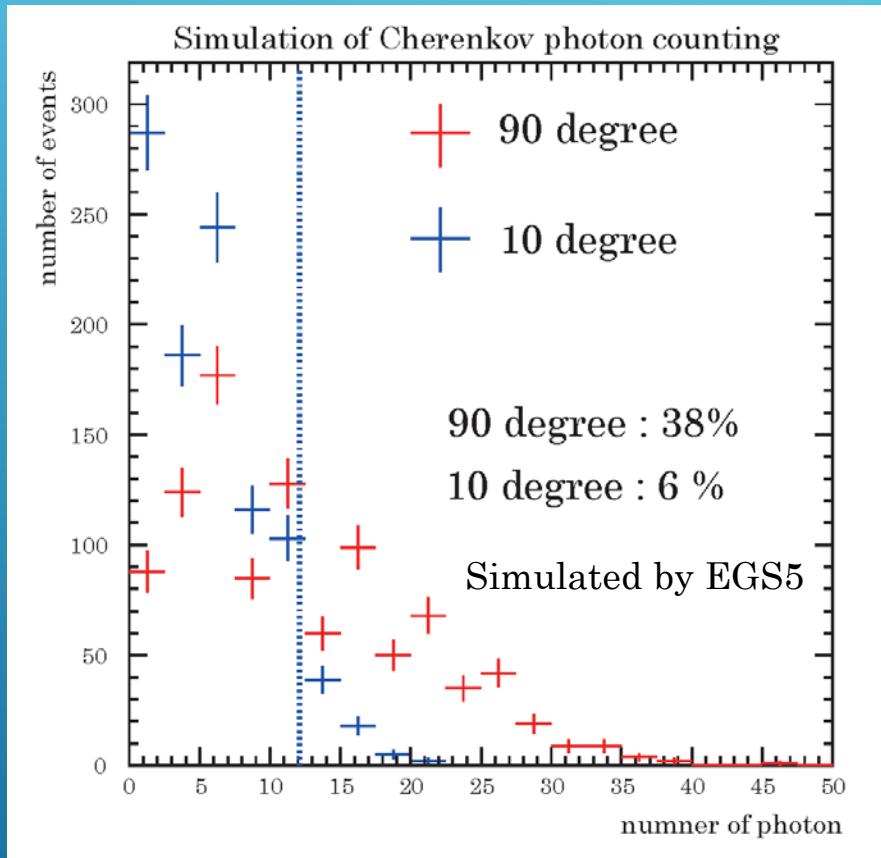
Decay branch of Thallium-208



Radiations	$y(i)$ $(\text{Bq}\cdot\text{s})^{-1}$
beta- 5	2.27×10^{-3}
beta- 8	3.09×10^{-2}
beta- 10	6.30×10^{-3}
beta- 11	2.45×10^{-1}
beta- 12	2.18×10^{-1}
beta- 13	4.87×10^{-1}
ce-K, gamma 3	4.04×10^{-3}
gamma 4	6.31×10^{-2}
ce-K, gamma 4	2.84×10^{-2}
ce-L, gamma 4	4.87×10^{-3}
gamma 6	2.26×10^{-1}
ce-K, gamma 6	1.97×10^{-2}
ce-L, gamma 6	3.32×10^{-3}
gamma 7	8.45×10^{-1}
ce-K, gamma 7	1.28×10^{-2}
ce-L, gamma 7	3.51×10^{-3}
gamma 13	1.81×10^{-2}
gamma 15	1.24×10^{-1}
ce-K, gamma 15	2.80×10^{-3}
gamma 19	3.97×10^{-3}
gamma 25	9.92×10^{-1}

The vertex reconstructed by scintillation make it within fiducial volume due to mis-fitting of gammas.

Monte Carlo Simulation



Number of event received Cherenkov light in Anisole has a clear difference between 90deg and 10deg, because of directionality of Cherenkov light.

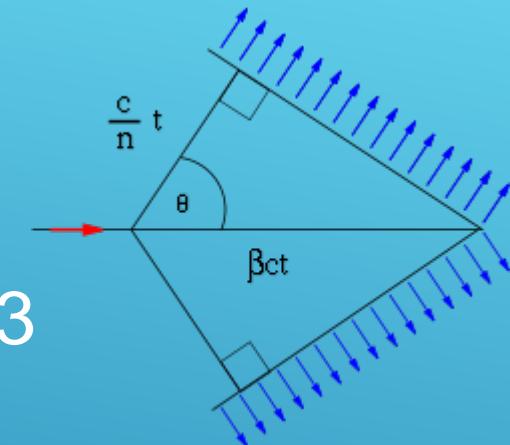


Maybe different situation in Liquid Scintillator.

Photon could be scattered by high concentration of solute.
We have to simulate such kind of effect.

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 angel $\theta=47.3$ degree are expected.
- Number of Cherenkov photon :
100 photon/MeV (400nm – 600nm)



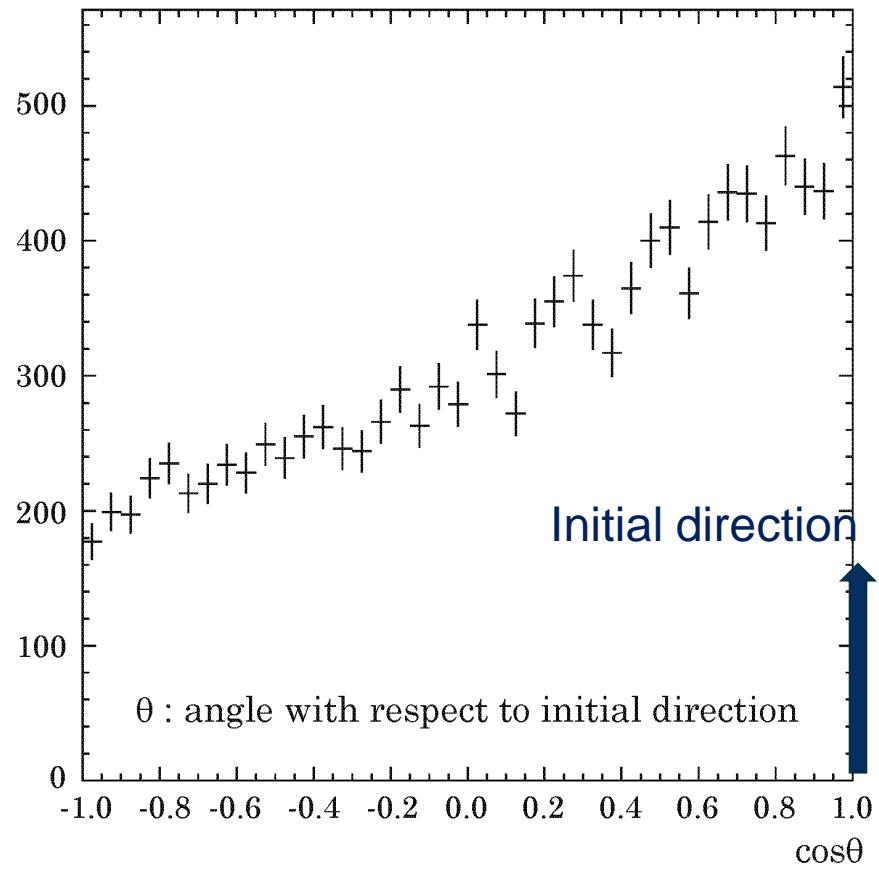
$$\frac{dN}{dx} = 2\pi z^2 \alpha \sin^2 \theta_c \int_{\lambda_1}^{\lambda_2} \frac{d\lambda}{\lambda} = 475 z^2 \sin^2 \theta_c \text{ photon/cm}$$

c.f. Light yield of Scintillation : $\sim 12000 \text{ photon/MeV}$

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

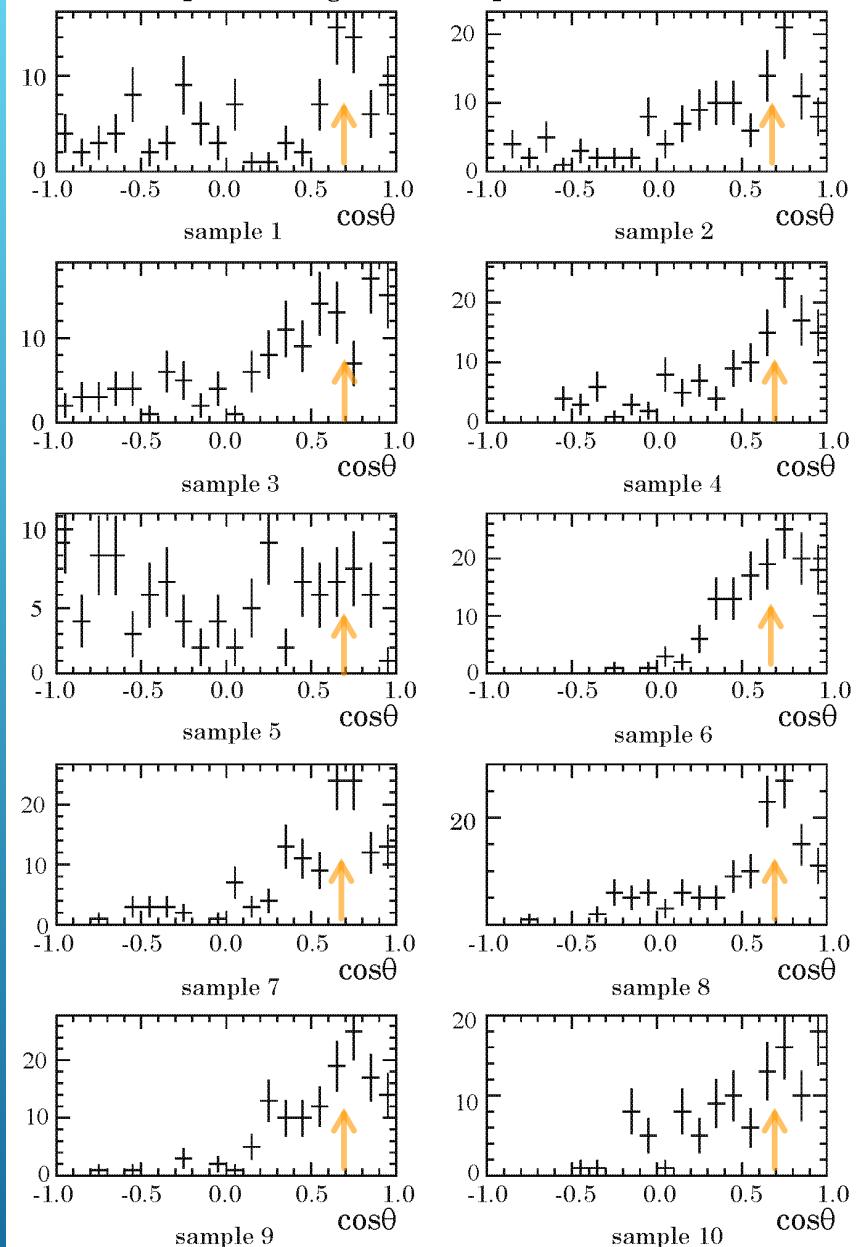
Multiple scattering

1.675MeV electron simulated by EGS5

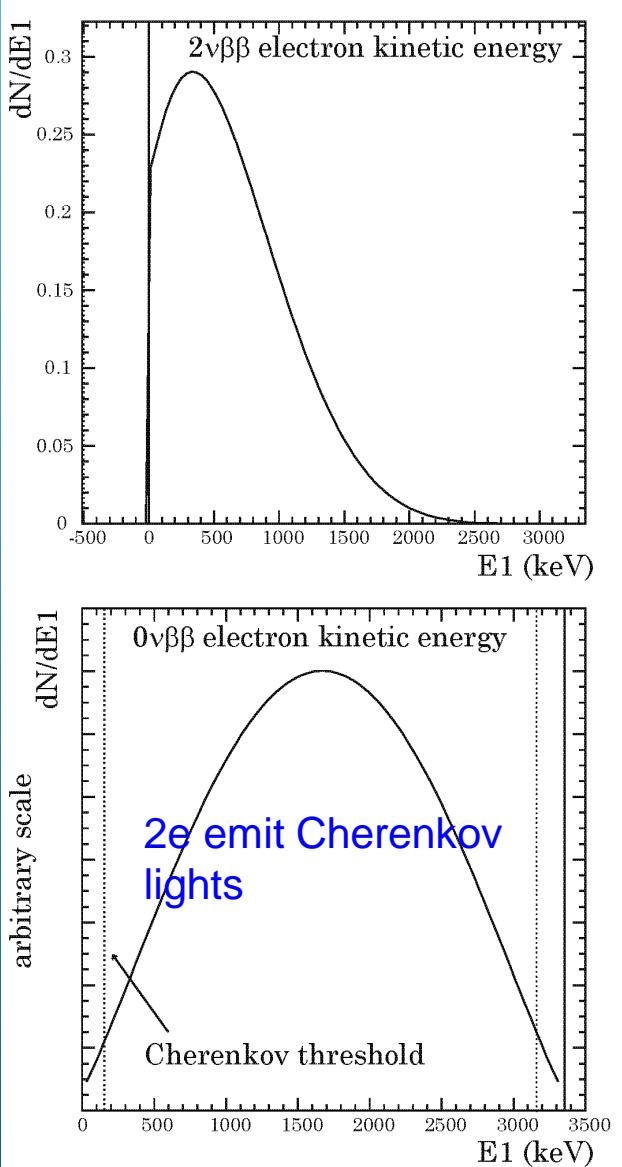


Even though multiple scattering of electrons, Cherenkov photons look have some clusters.

Cherenkov photon angle with respect to the initial direction



kinetic energy spectrum of electron



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

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

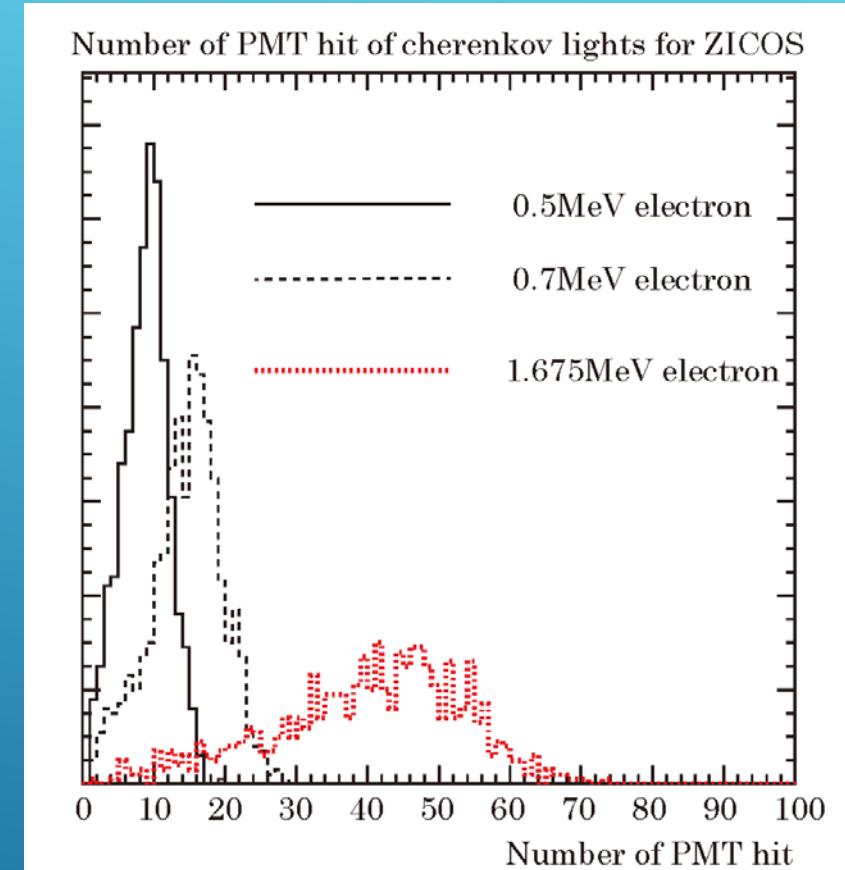
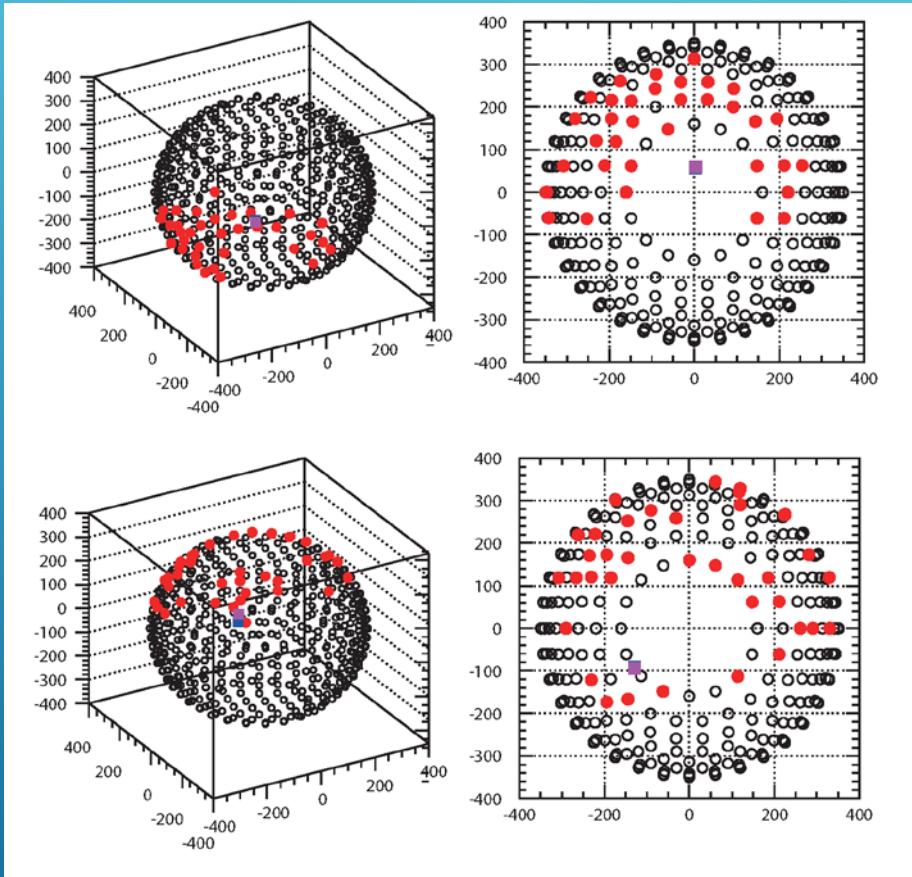
ε_i can generate independently within energy conservation.

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

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

Simulation of Cherenkov lights

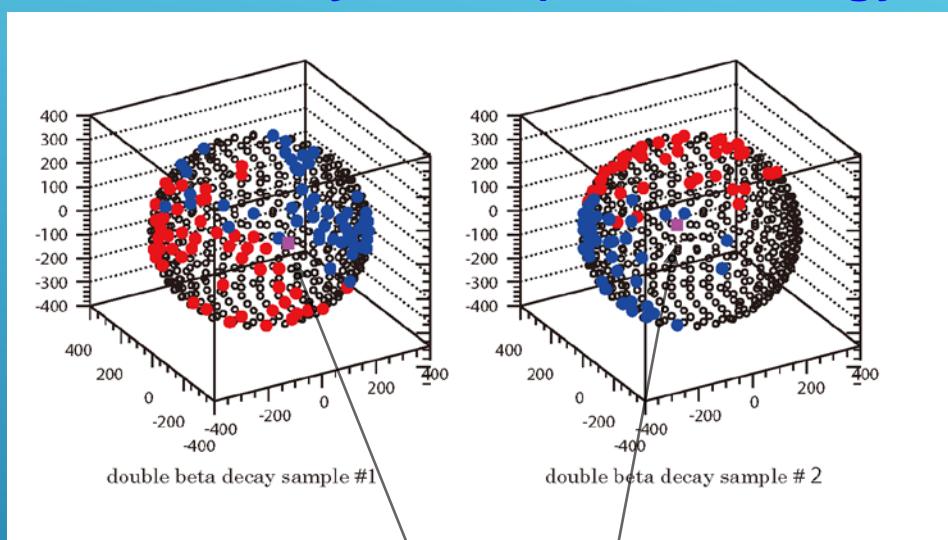
Simulated by EGS5 (kinetic energy 1.675MeV)



Hard to detect Cherenkov events below 0.5MeV.

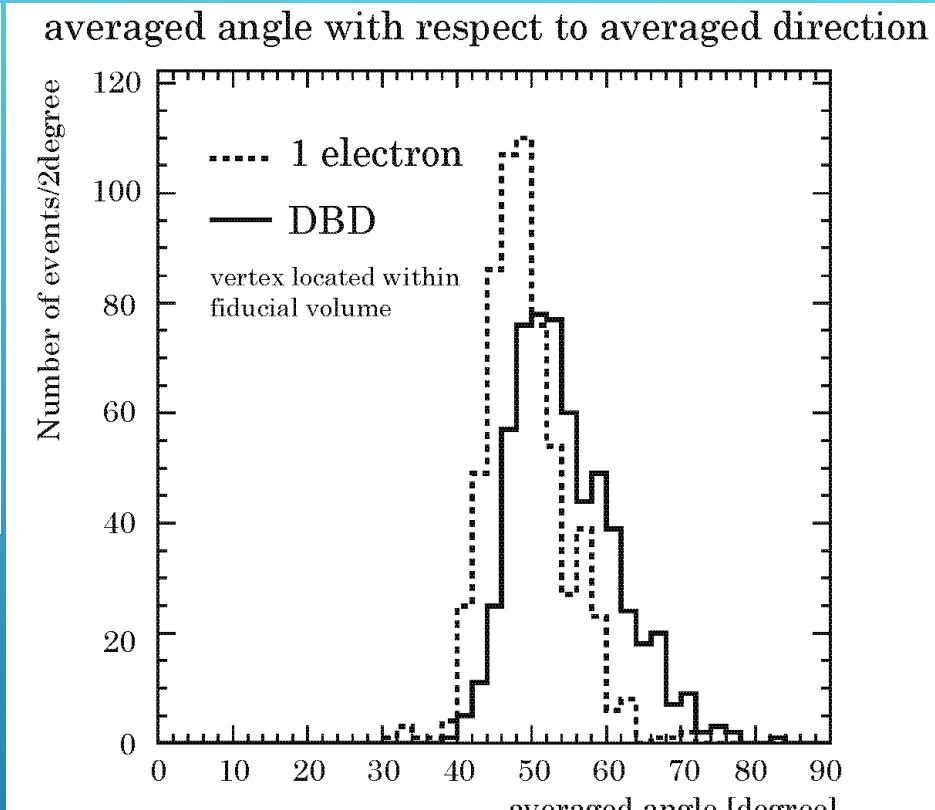
Hit pattern of DBD (opposite and half E)

Simulated by EGS5 (kinetic energy 1.675MeV)

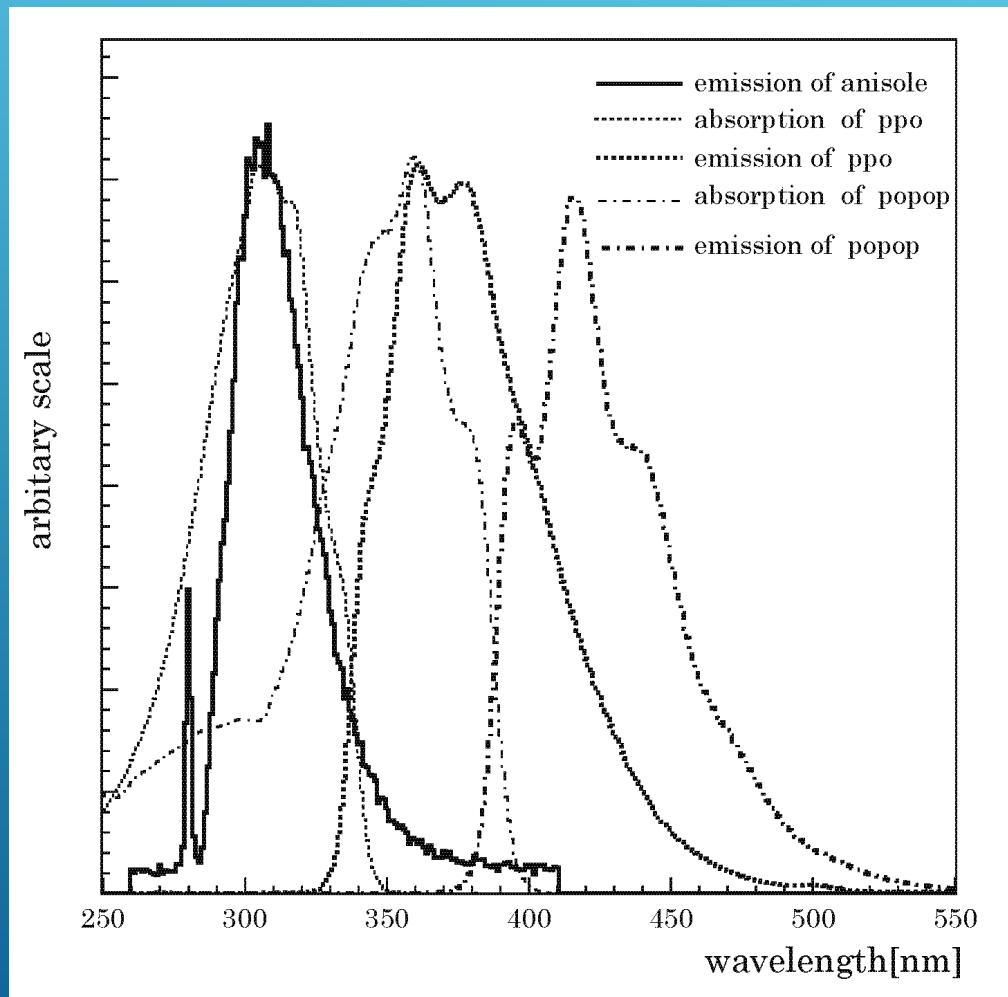


Generate position of DBD

Multi events from DBD
tend to have a slightly
larger values of averaged
angle than single e⁻.



Emission and absorption spectra for solvent and solute in standard cocktail

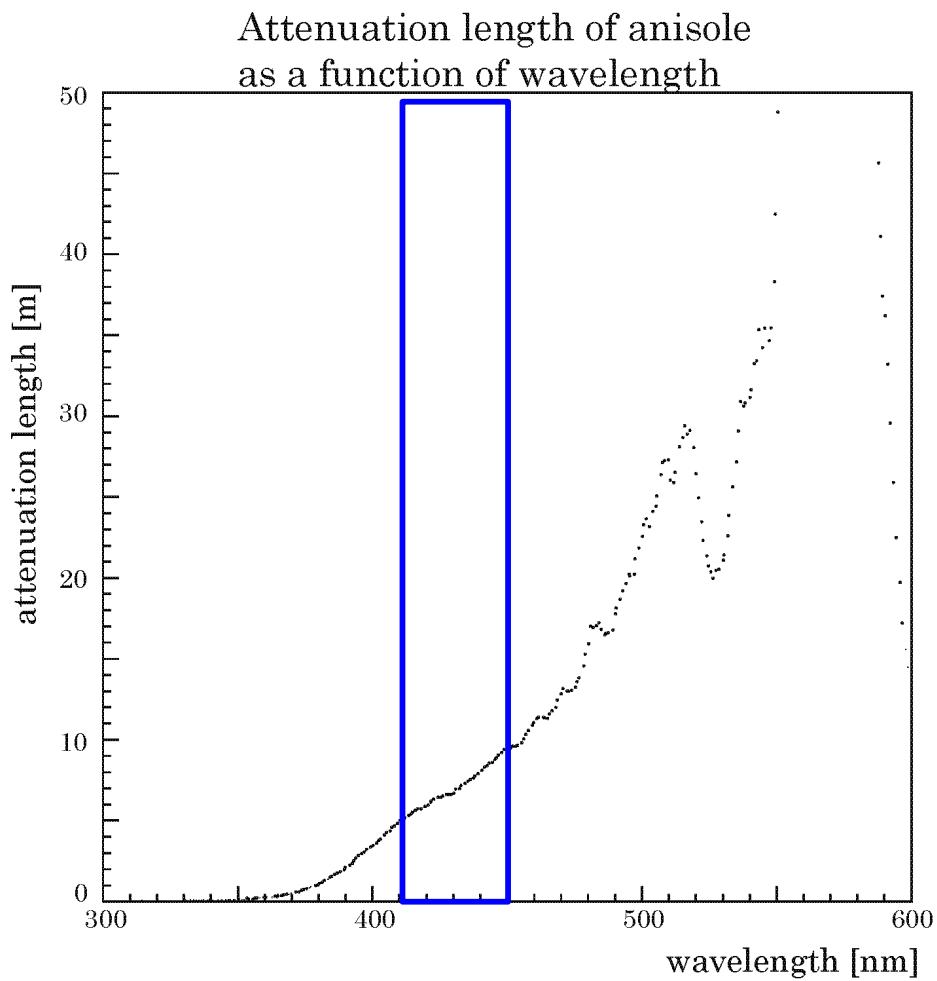


PPO absorbed most
of emission lights from
anisole.



Effectively the energy
was transferred to the
secondary scintillator.

ATTENUATION LENGTH OF ANISOLE

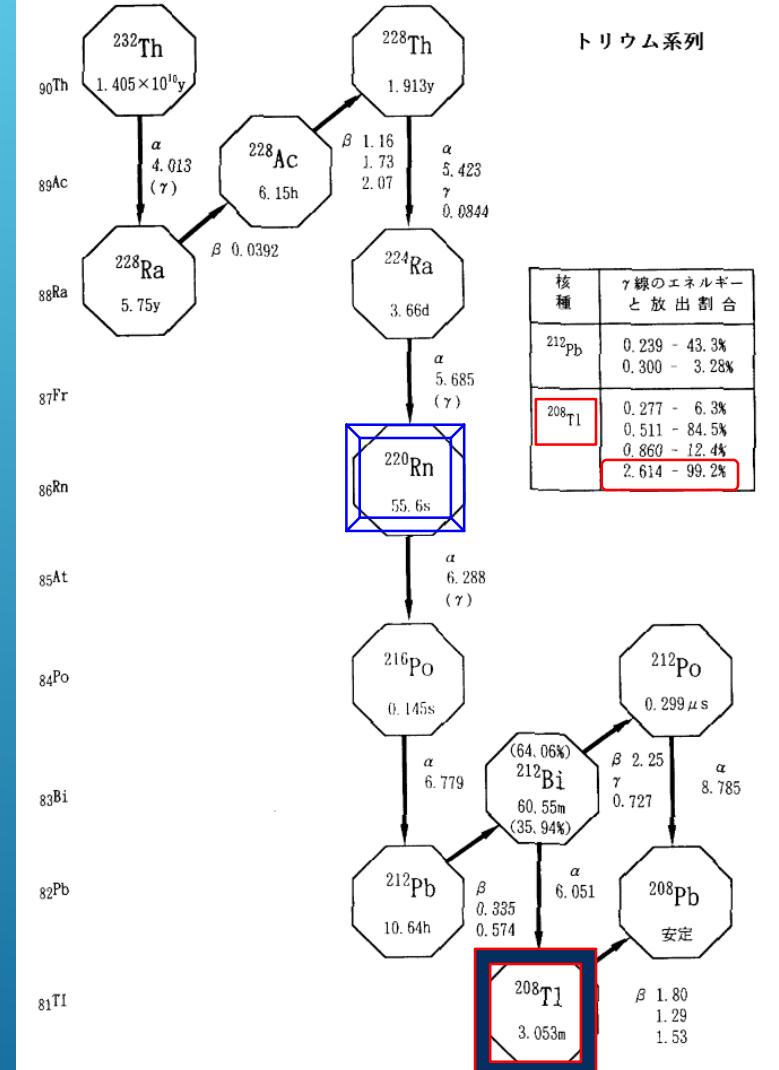
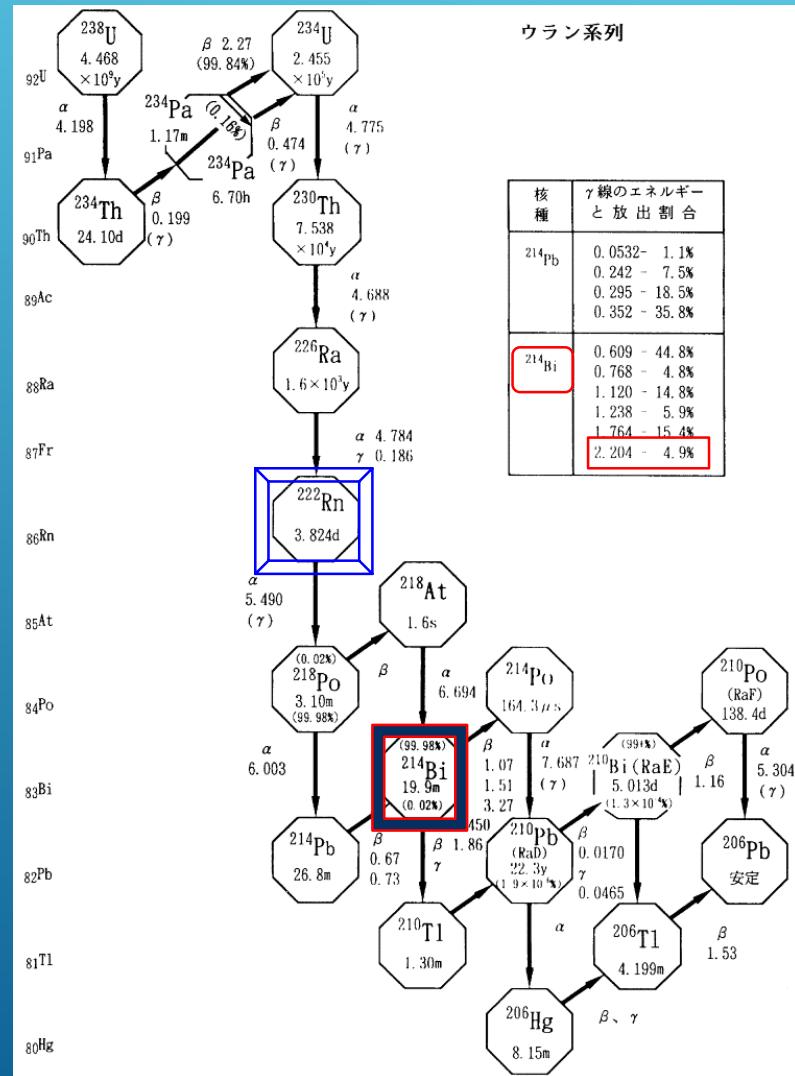


Attenuation length of scintillation light from POPOP ($\sim 450\text{nm}$) was obtained as $\sim 6\text{m}$.

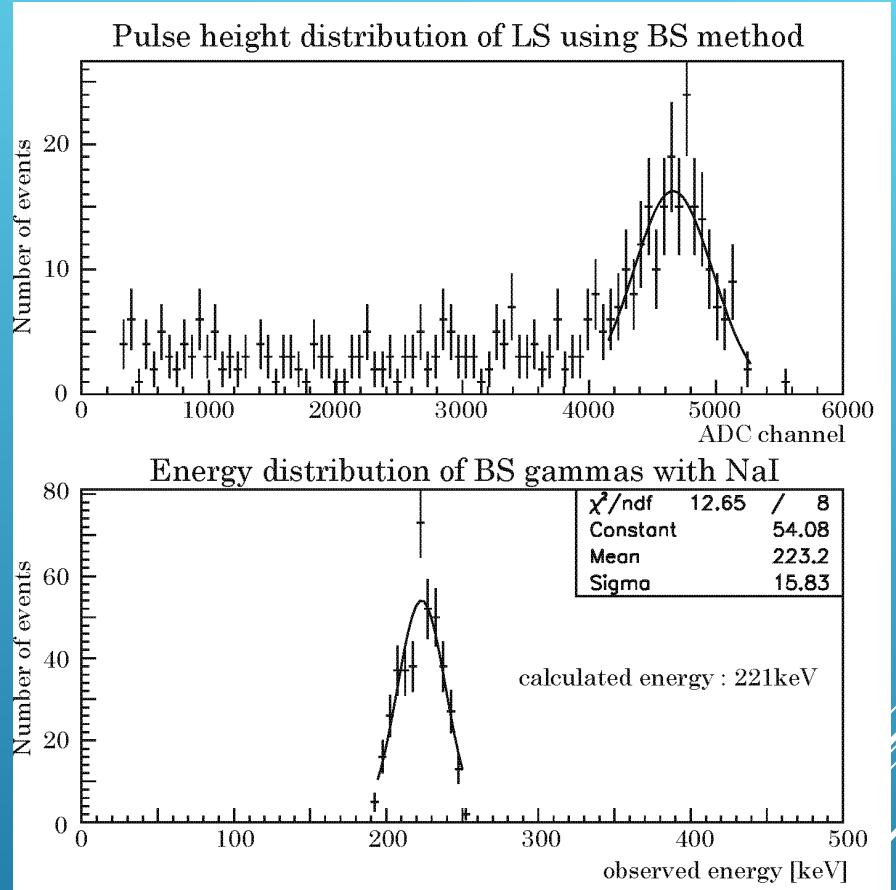
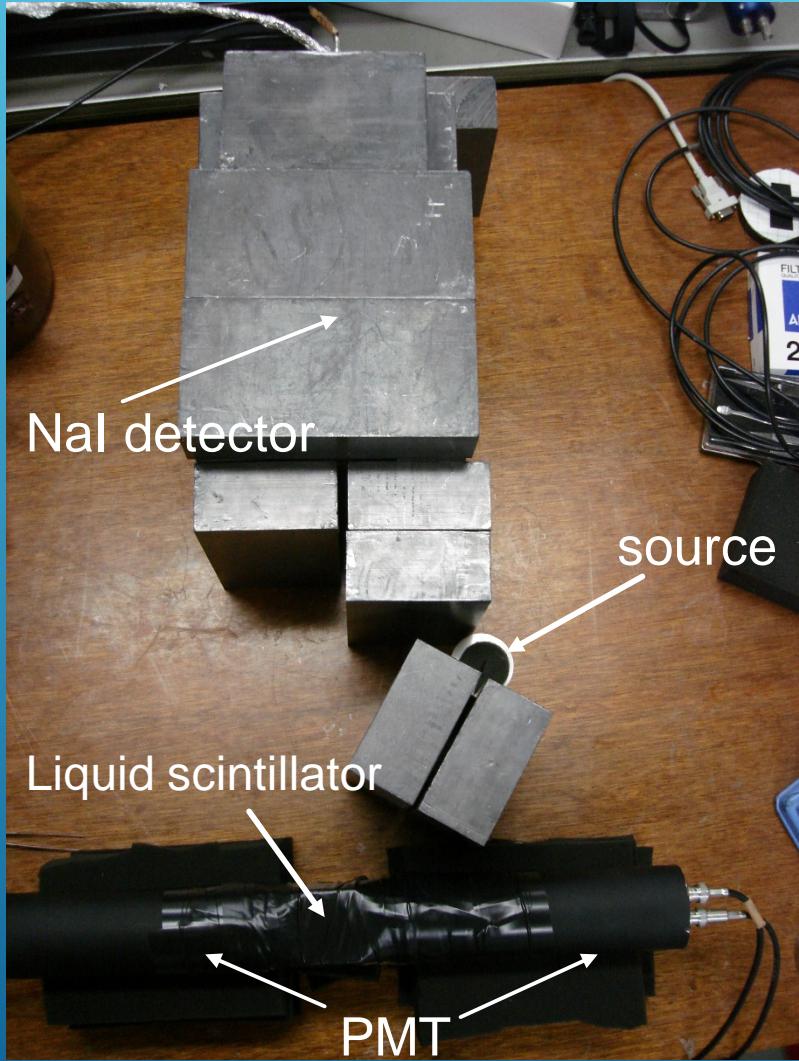


No problem for radius of ZICOS detector.

Natural radiative U/Th decay chain

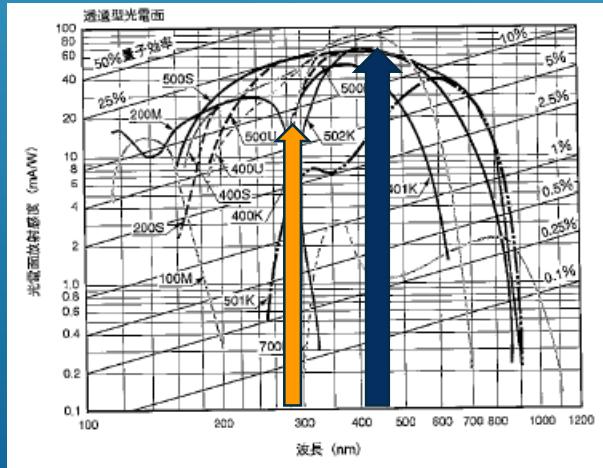
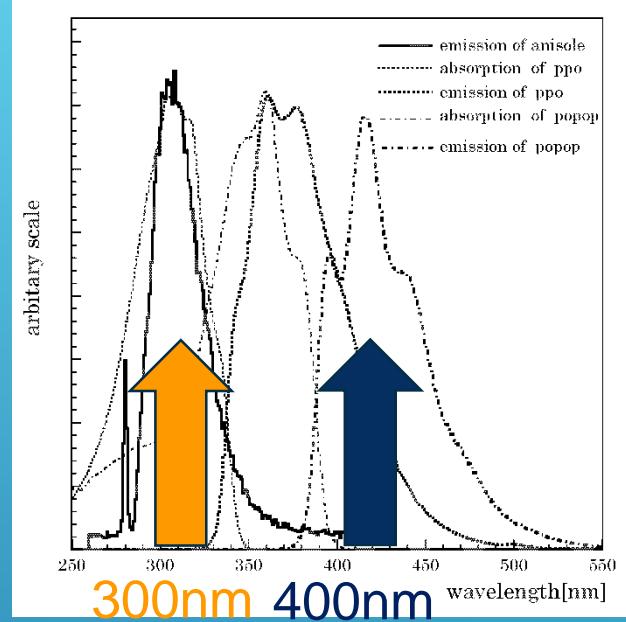
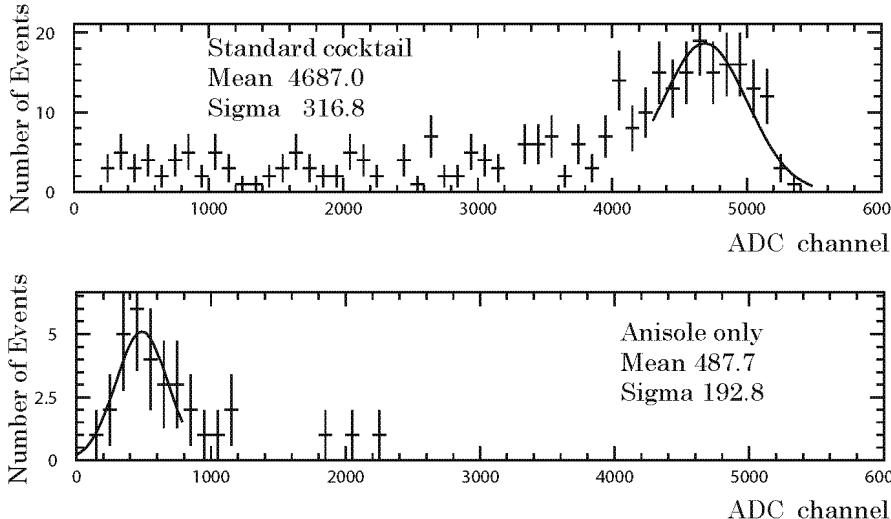


Backscattering method



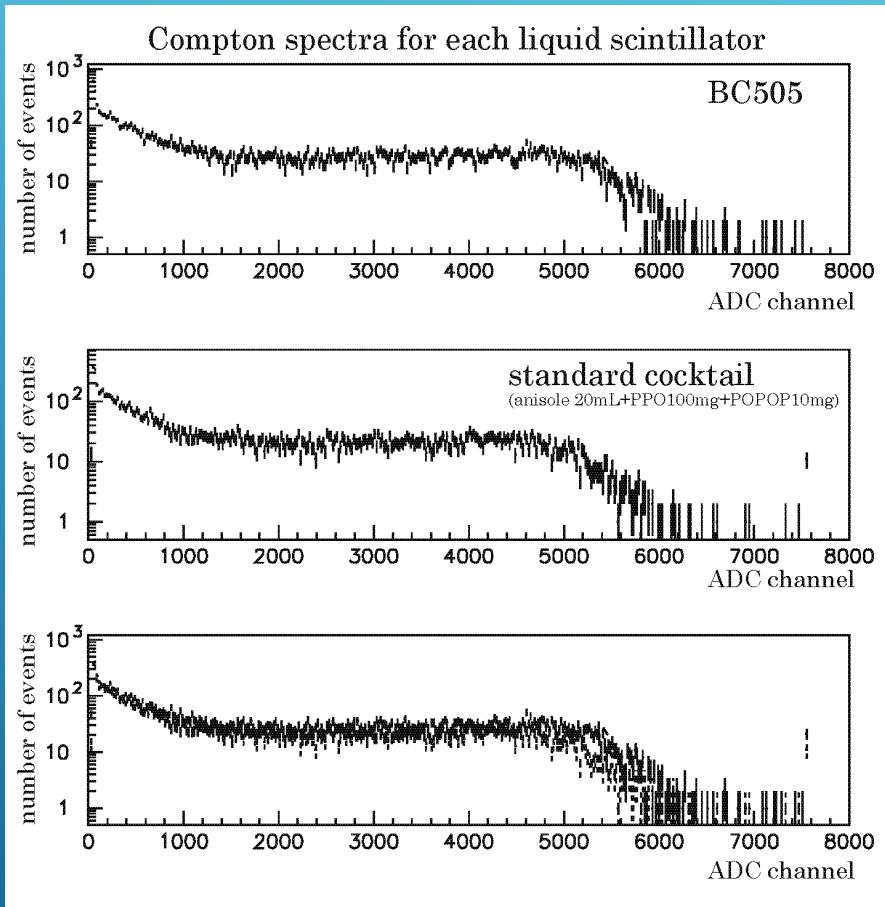
Single peak could be used even in liquid scintillator.

Light yield of scintillation in anisole



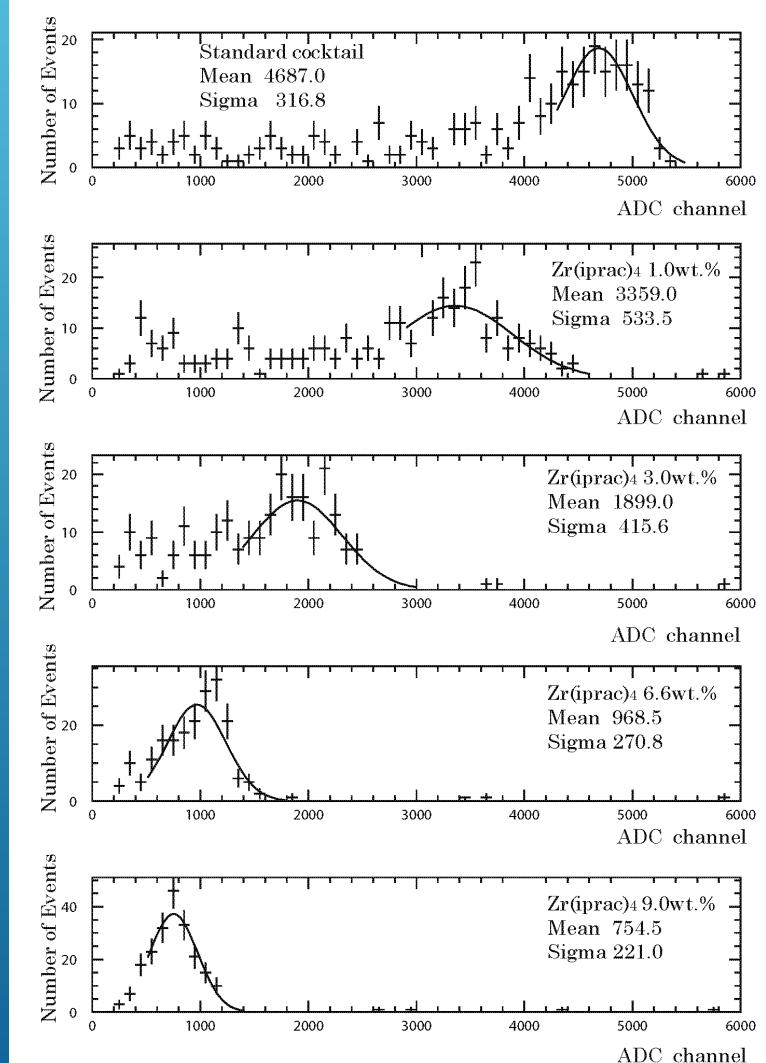
Relative scintillation light yield of **anisole** is 9.8% to **standard cocktail** (due to difference of quantum efficiency of PMT)

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)₄.

Energy resolutions are also getting worth as a function of the concentration of Zr(iprac)₄.

Physical constants of Liquid Scintillator

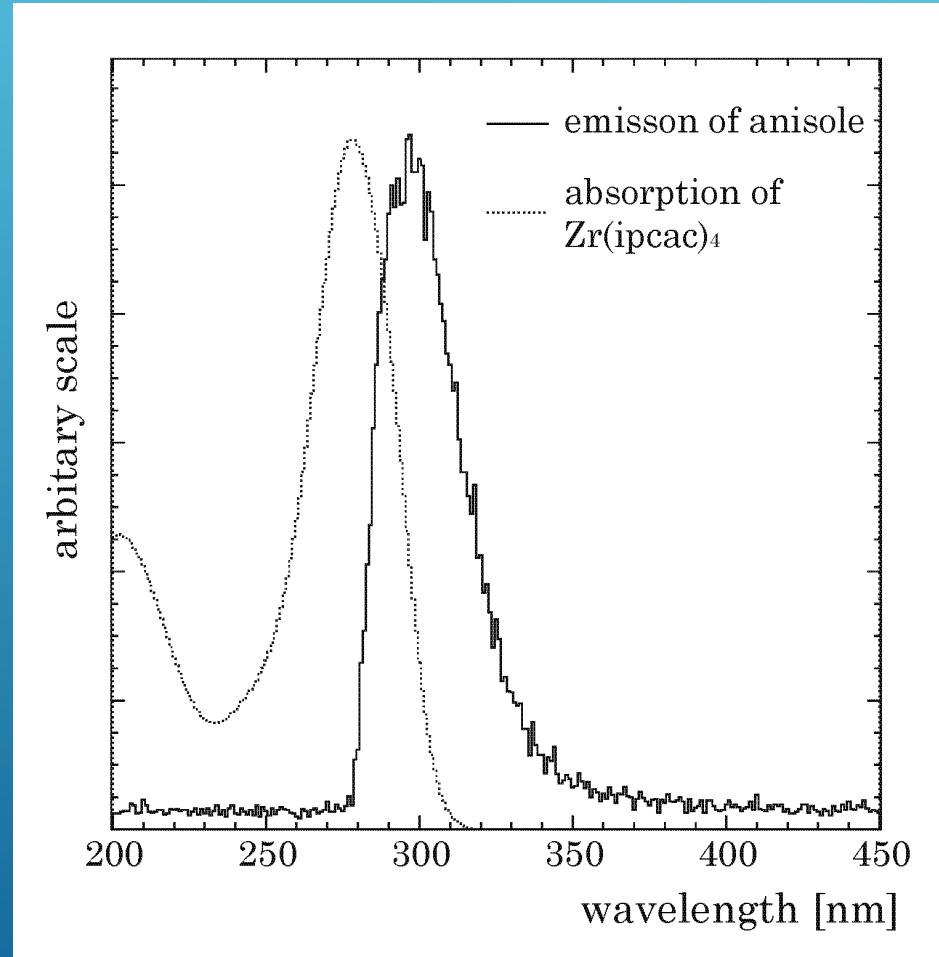
Physical Constants of SGC Liquid Scintillators

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

Absorbance spectra for Zr(iprac)_4

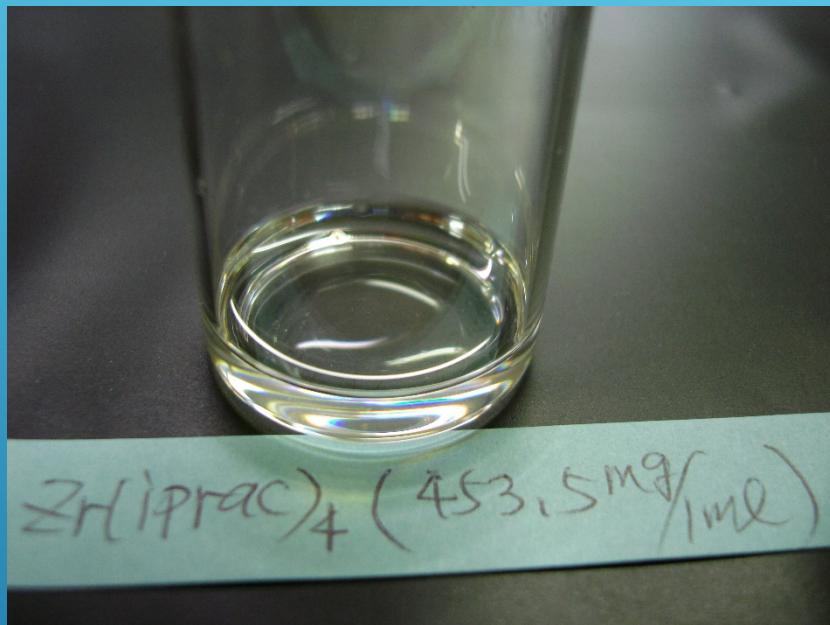


Absorption peaks of Zr(iprac)_4 was found around at 278nm. However, overlapped region with emission of anisole was existed.



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

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.

Light yield quenching by Zr(iprac)₄

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

L_0 : Light yield of anisole

N_{ppo} : Number of PPO molecular in mole

N_{Zr} : Number Zr complex molecular in mole

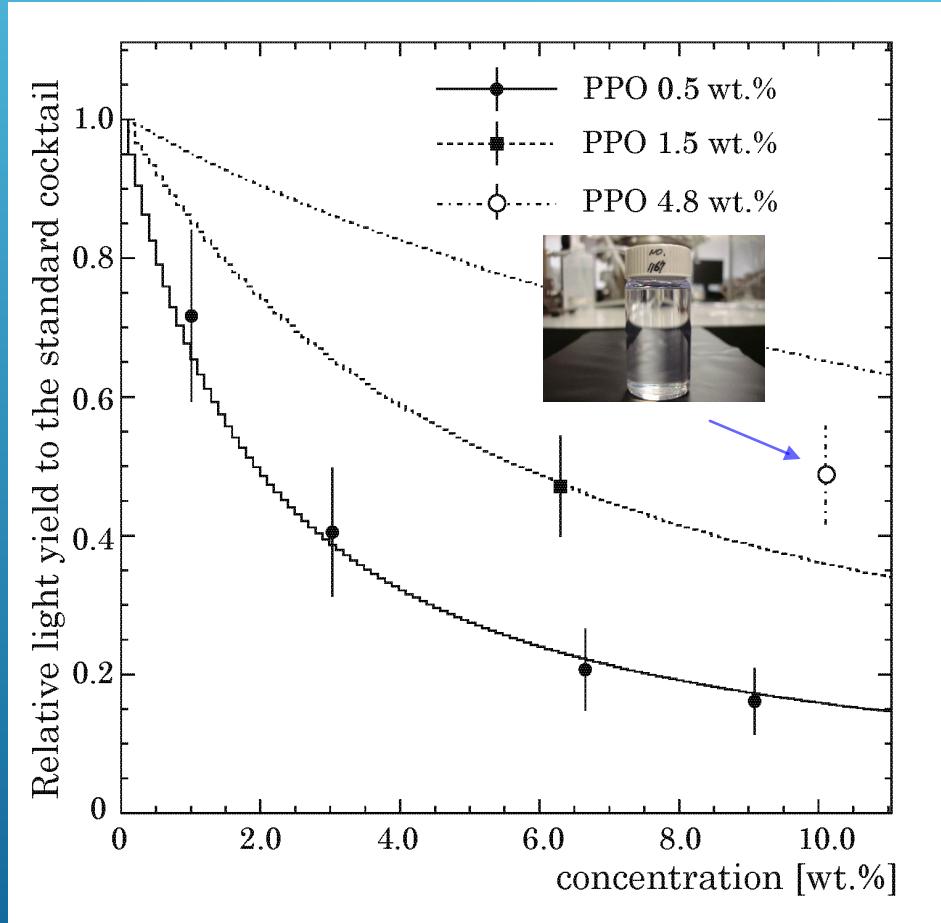
σ_1 : absorbance of PPO (mol⁻¹)

σ_2 : absorbance of Zr complex (mol⁻¹)

PPO would help the recovering light yield.

Recovering the light yield

Measured at several conditions of PPO concentration



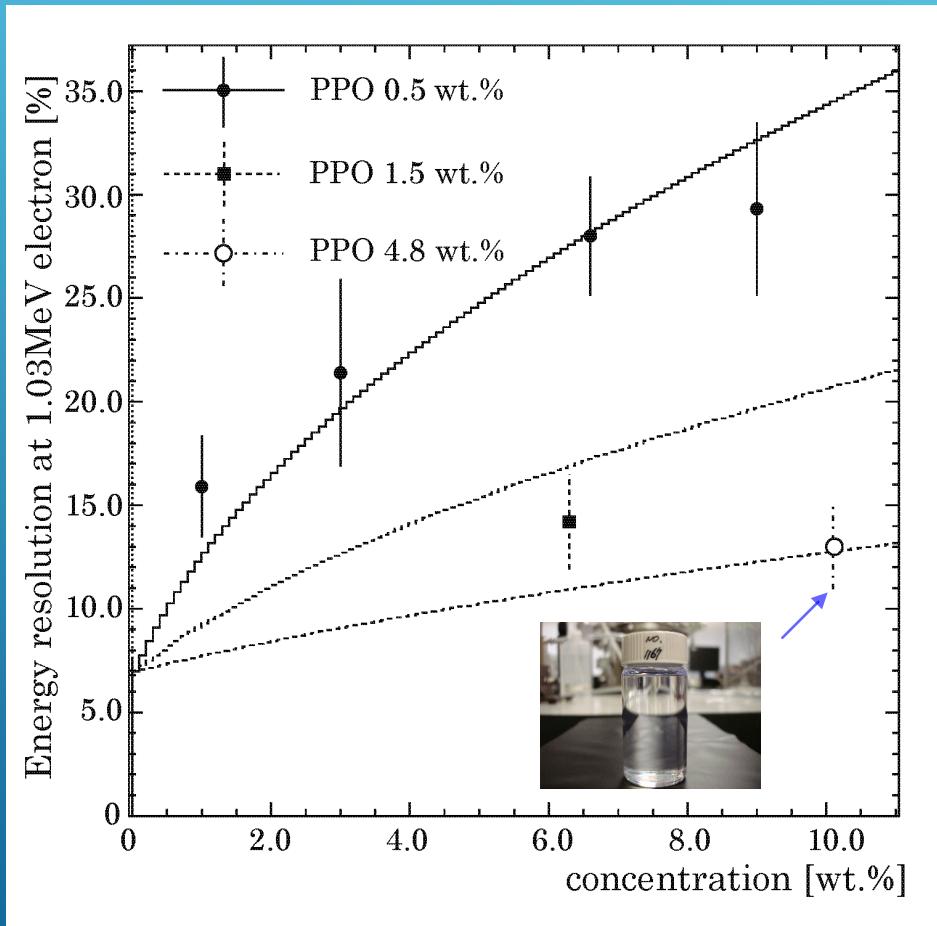
5wt.% PPO helps
actually recovering the
scintillation light yield.



**$48.7 \pm 7.1\%$ light yield
to standard cocktail
was obtained at
10wt.% concentration.**

Recovering the energy resolution

Measured at several conditions of PPO concentration

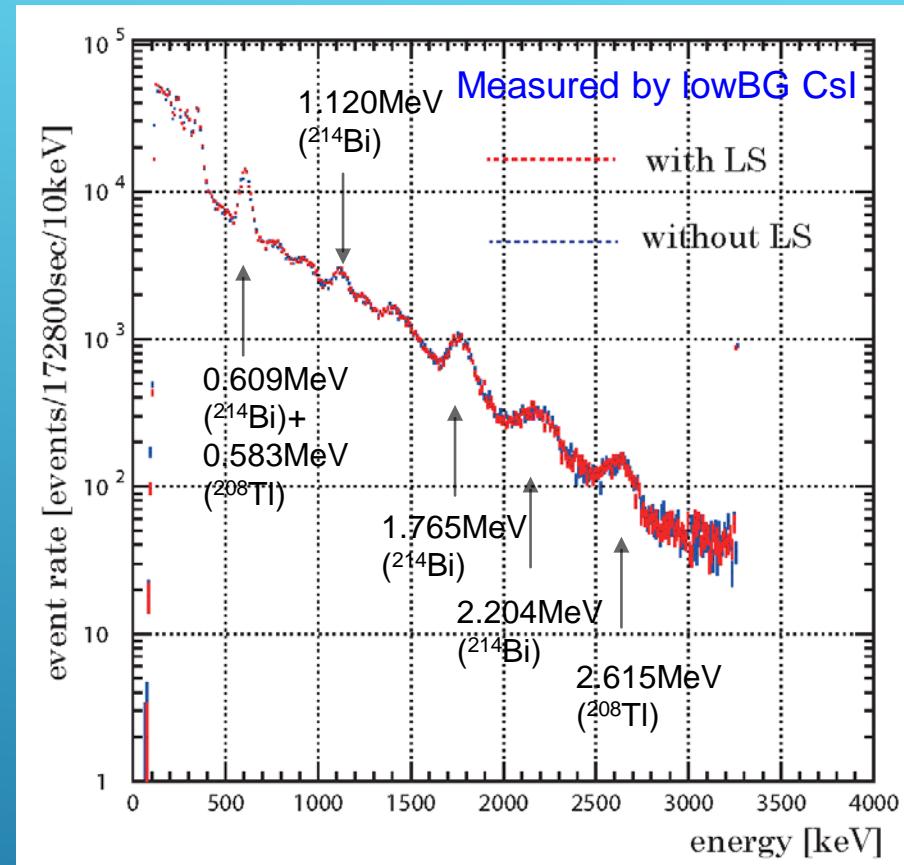


5wt.% PPO helps again
the energy resolution
35% → 13%. at 10wt.%
of Zr(iprac)₄.

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

Achieved goal !

Measurement of backgrounds from LS



Using subtracted # of events around 2.6MeV and 2.2MeV

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

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