

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

TAUP 2017 XV International Conference on
Topics in Astroparticle and Underground Physics

Sudbury Canada

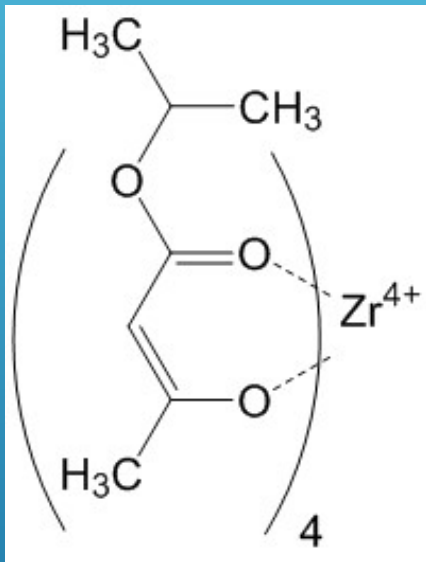
25 July, 2017

Miyagi University of Education Y. Fukuda, Narengerile, A.Obata,
Y.Kamei

Kamioka Observatory, ICRR, Univ. of Tokyo S. Moriyama
Fukui University I. Ogawa

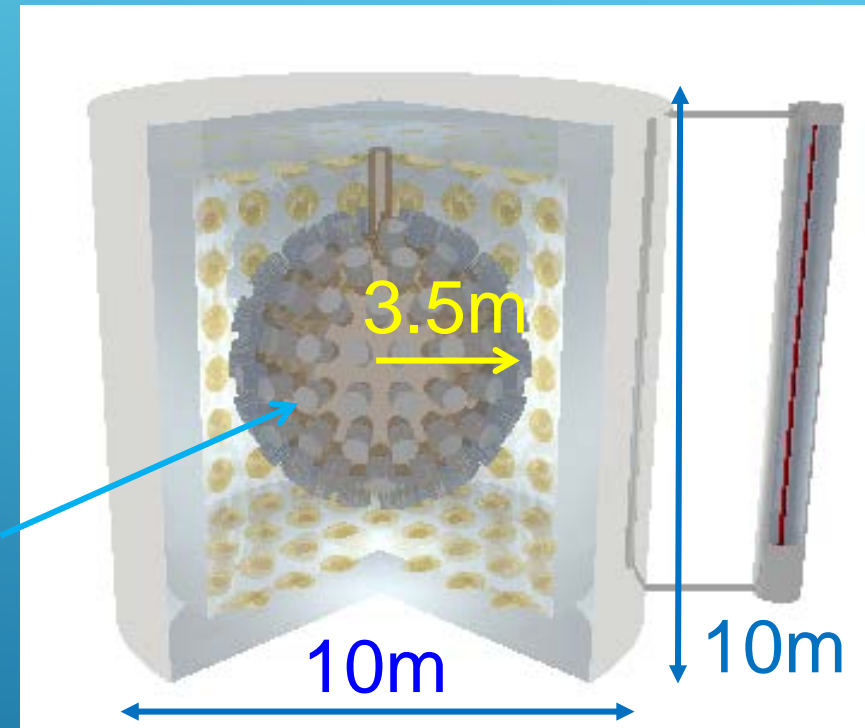
Tokyo University of Science T. Gunji, S. Tsukada, R. Hayami

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



Tetrakis(iso-propyl acetoacetate) zirconium : $Zr(iprac)_4$
MW : 663.87

LS: $Zr(iprac)_4$ 10wt.% and PPO 5 wt.% solved in anisole.



Estimated energy resolution $\sim 2.8\%$ @ 3.35MeV assuming 64% photo coverage.

Neutrino mass sensitivity of ZICOS experiment

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

Measurement time: 2years

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 ⁹⁶Zr (e.g. 57.3% for NEMO-3)

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

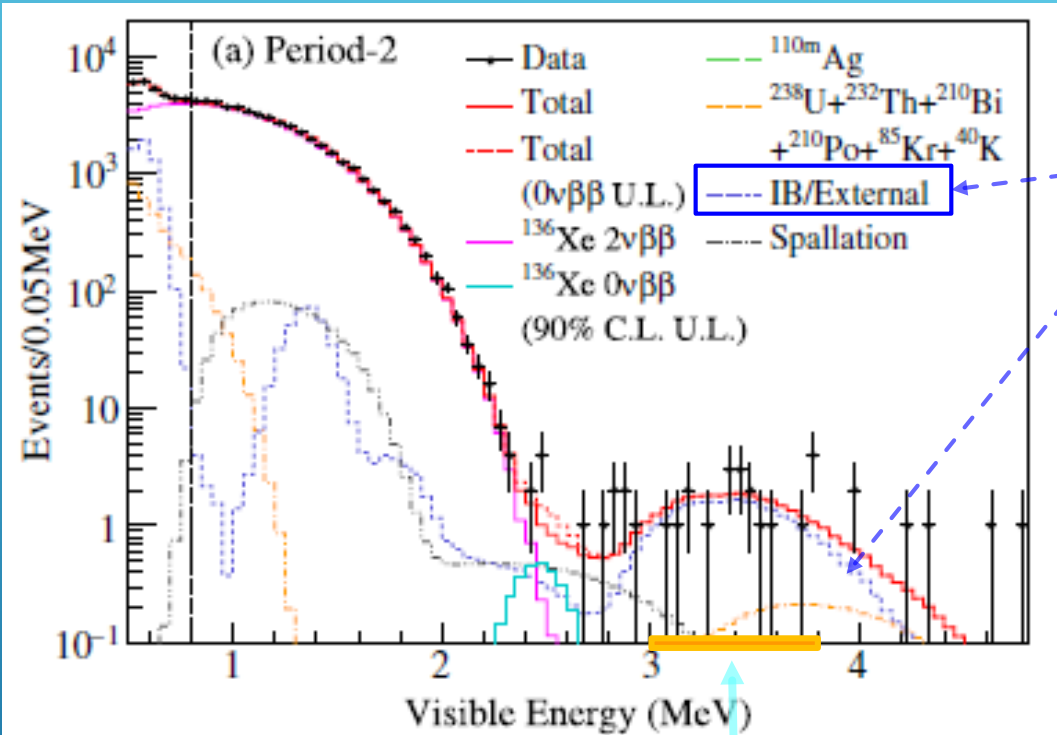
2) BG (²⁰⁸Tl) reduction

BG level < 1/20 × KL-Zen

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

Backgrounds around signal region

Measured by KamLAND-Zen



^{208}Tl on surface of balloon

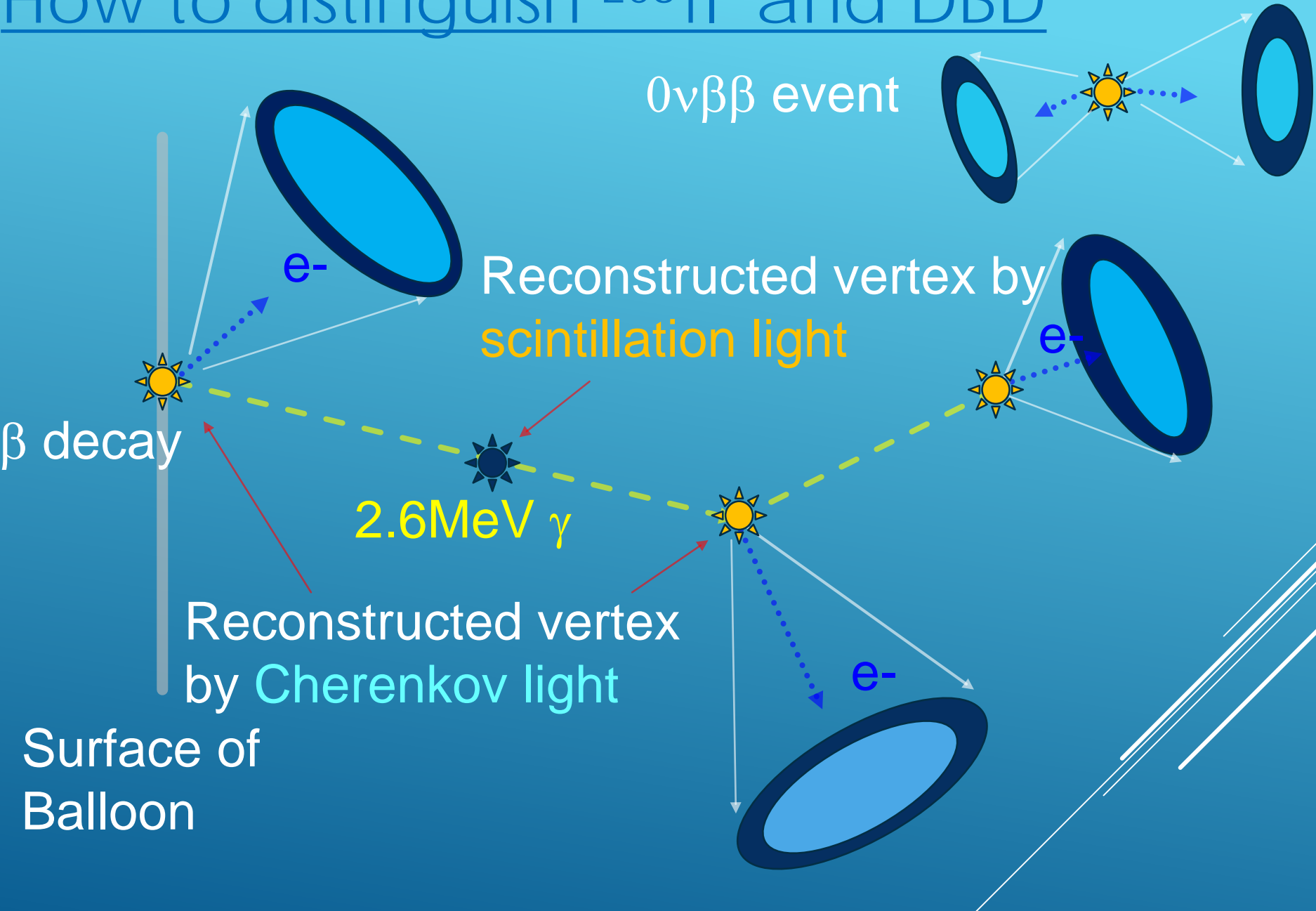
Necessary same clean material as KamLAND-Zen, but ... need further BG (^{208}Tl) reduction

Phys.Rev.Lett. 117 (2016) 082503

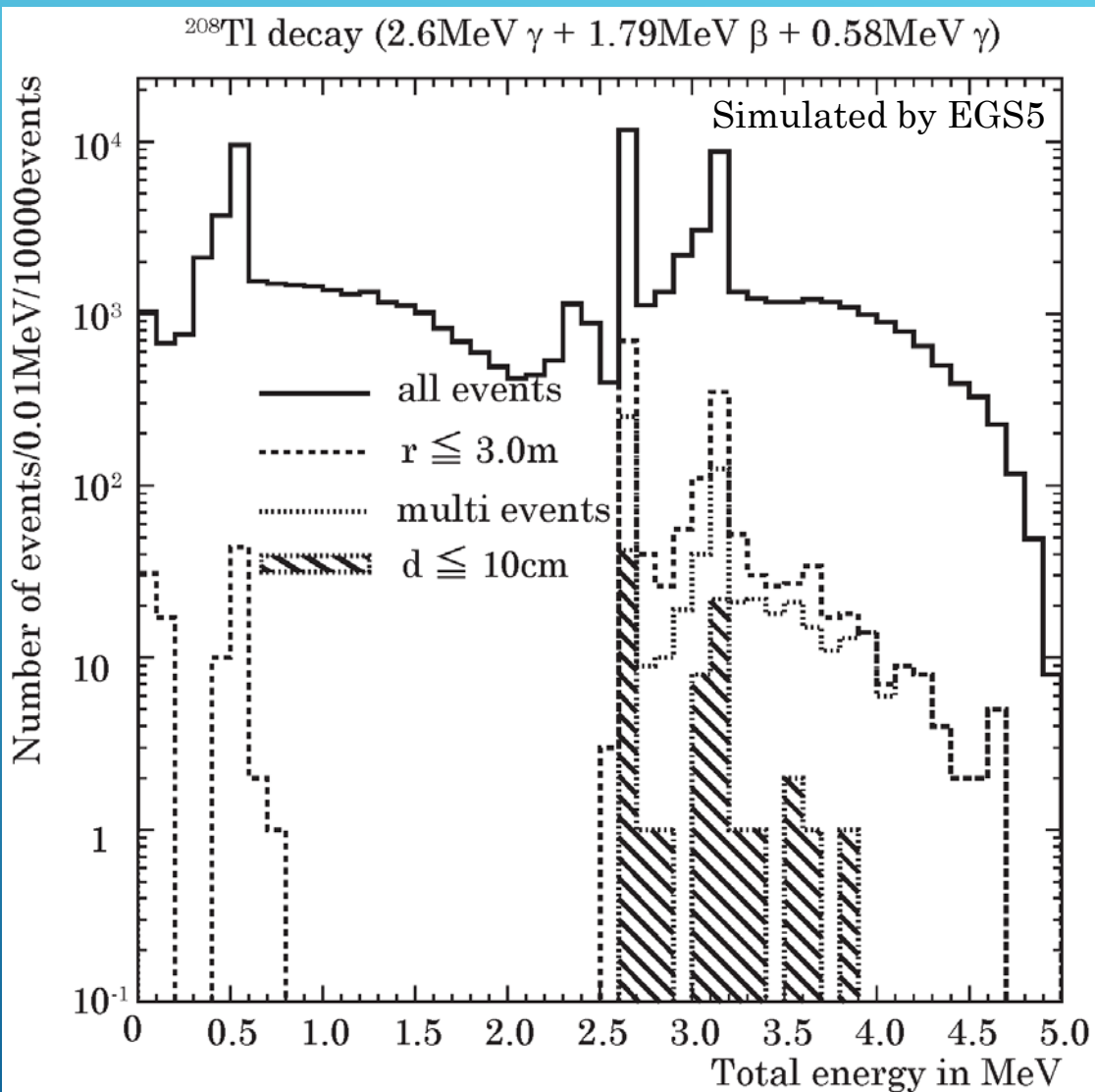
$0\nu\beta\beta$ signal region for ^{96}Zr

Require an additional technique other than the energy spectral shape obtained by scintillation.

How to distinguish ^{208}Tl and DBD



Reduction of ^{208}Tl decay

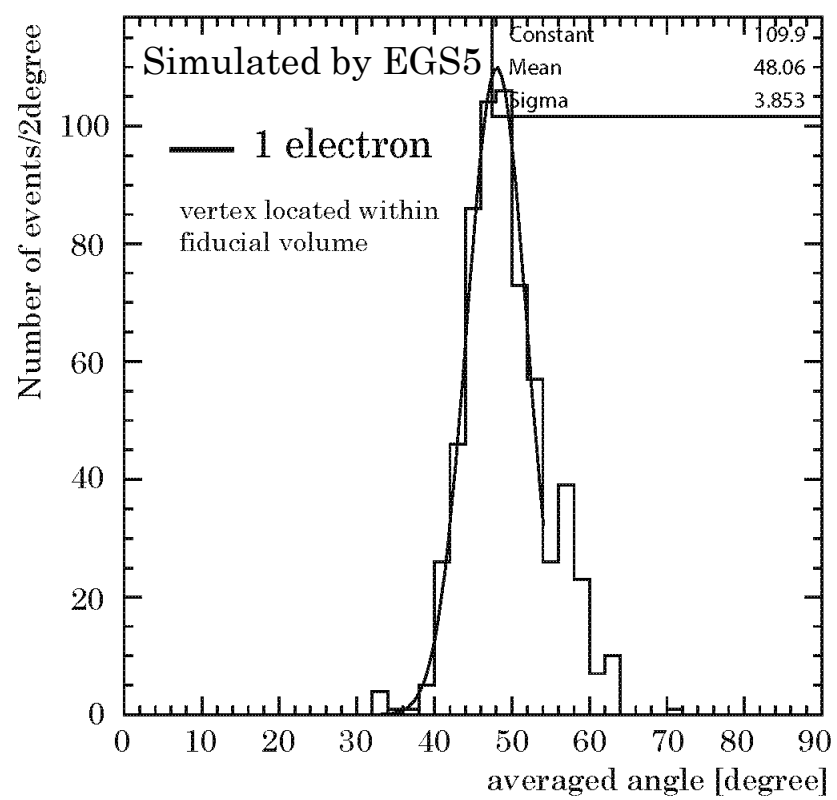


- 1) $E : 3.0\text{-}3.7\text{MeV}$
17925 events
- 2) Fiducial volume
628 events
- 3) Multi events
263 events
- 4) Closer events
($d \leq 10\text{cm}$)
35 events

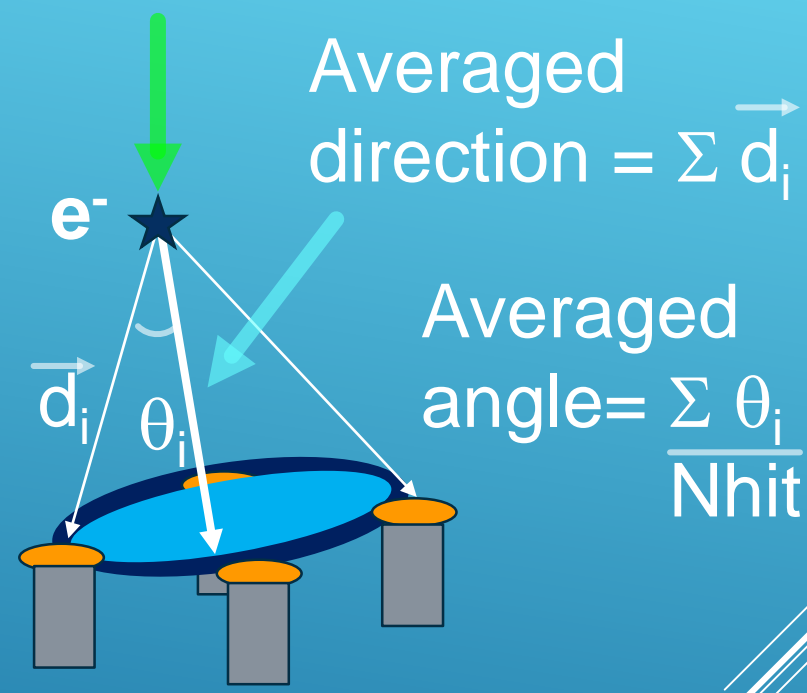
**~1/20 BG reduction
could be achieved by
using the information
from Cherenkov light.**

Averaged angle of Cherenkov hit

averaged angle with respect to averaged direction



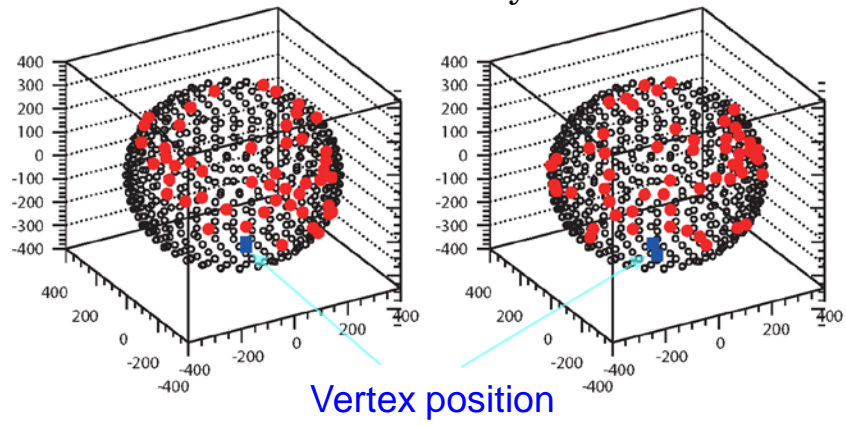
Vertex position obtained by scintillation



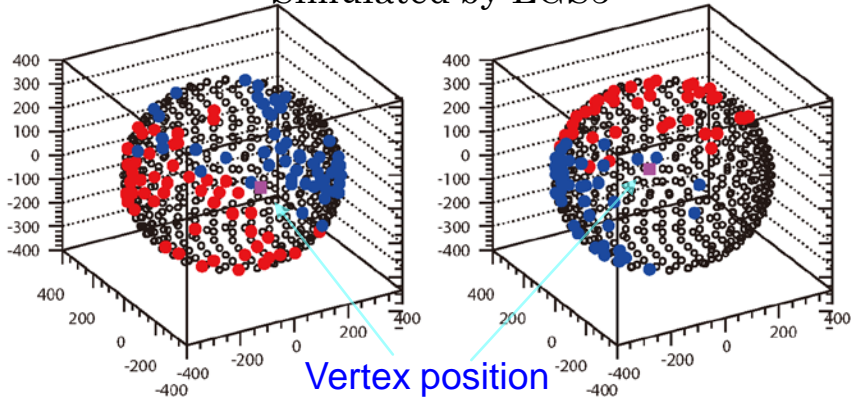
Averaged angle distribution with respect to averaged direction for single electron has a peak at ~48 degree, which is almost same as Cherenkov angle.

Hit pattern of ^{208}Tl decay and DBD

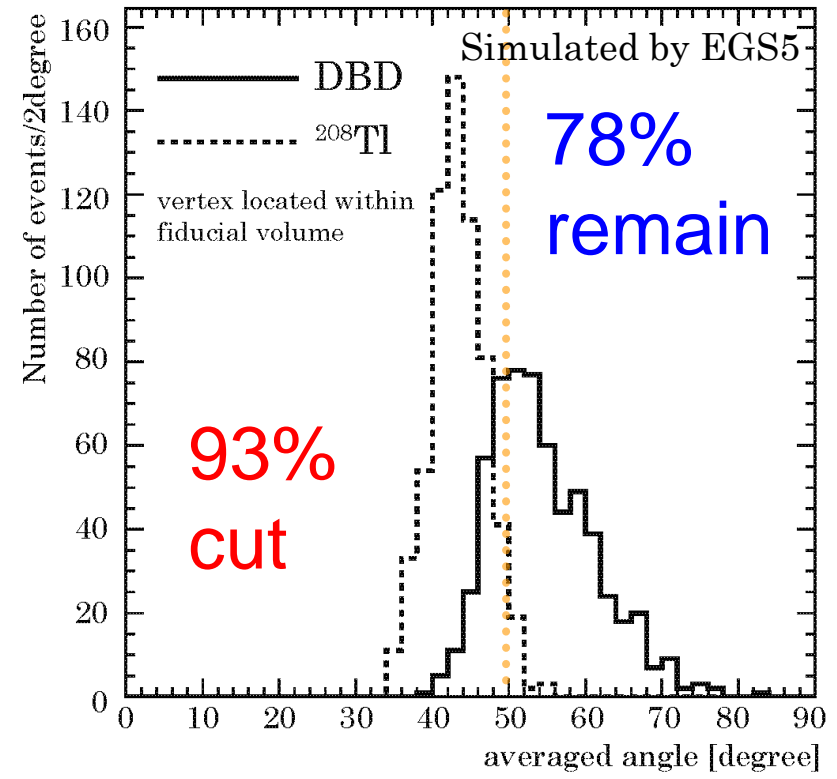
^{208}Tl beta-gamma multi events sample
Simulated by EGS5



Double beta decay event sample
Simulated by EGS5



averaged angle with respect to averaged direction

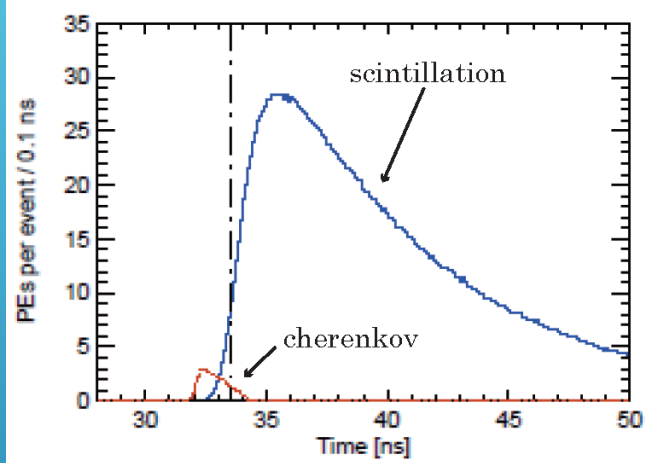


Hit pattern of Cherenkov lights for ^{208}Tl decay looks different from DBD.

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

Separation of Cherenkov and Scintillation

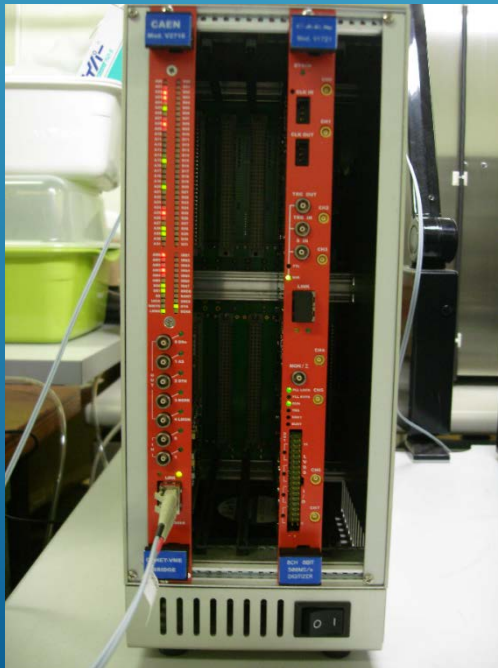
arXiv:1609.0986(simulation)



- Rise time of Cherenkov light : an order of a few 100 pico second due to the electromagnetic process
- Rise time of Scintillation light: an order of nano seconds in general.

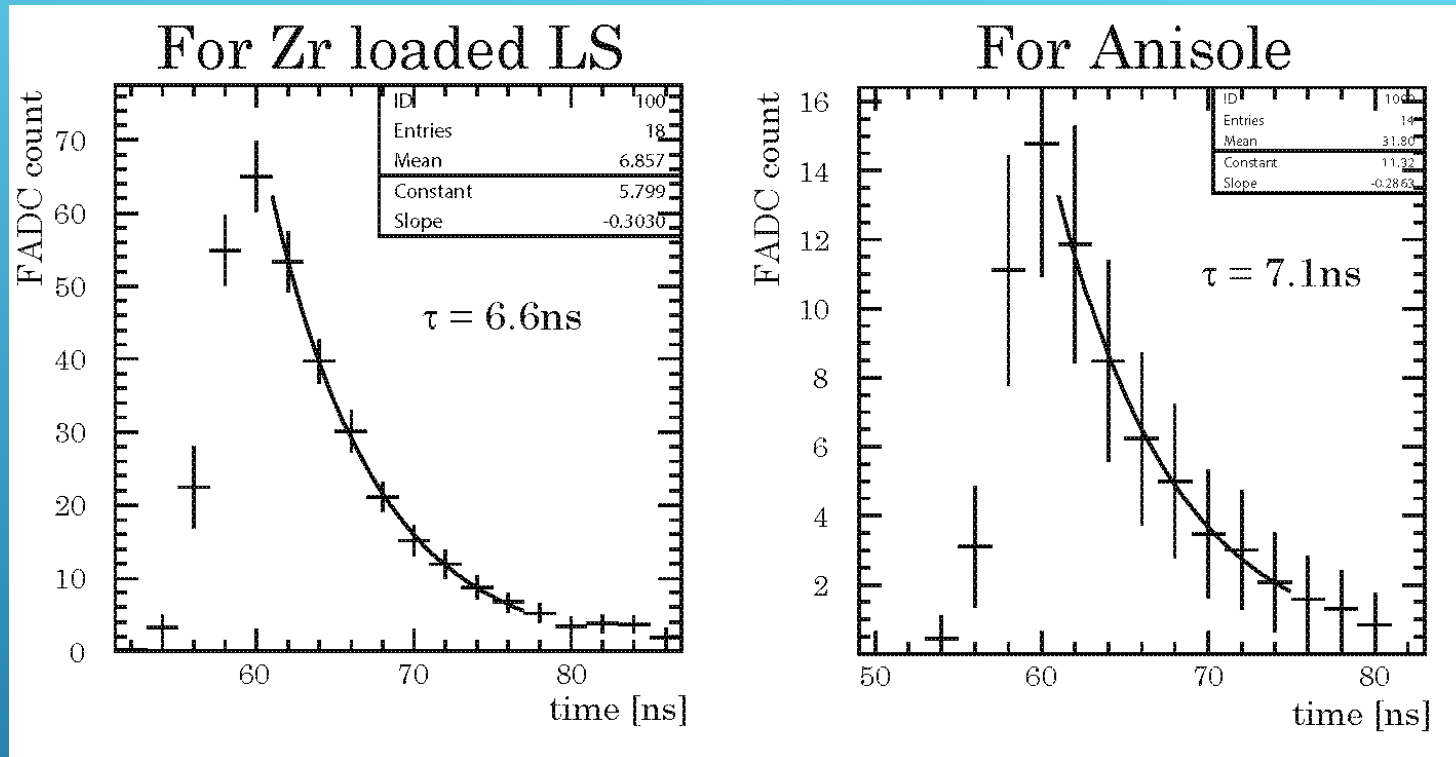


Possible to extract PMT hits received Cherenkov lights by **Pulse Shape Discrimination.**



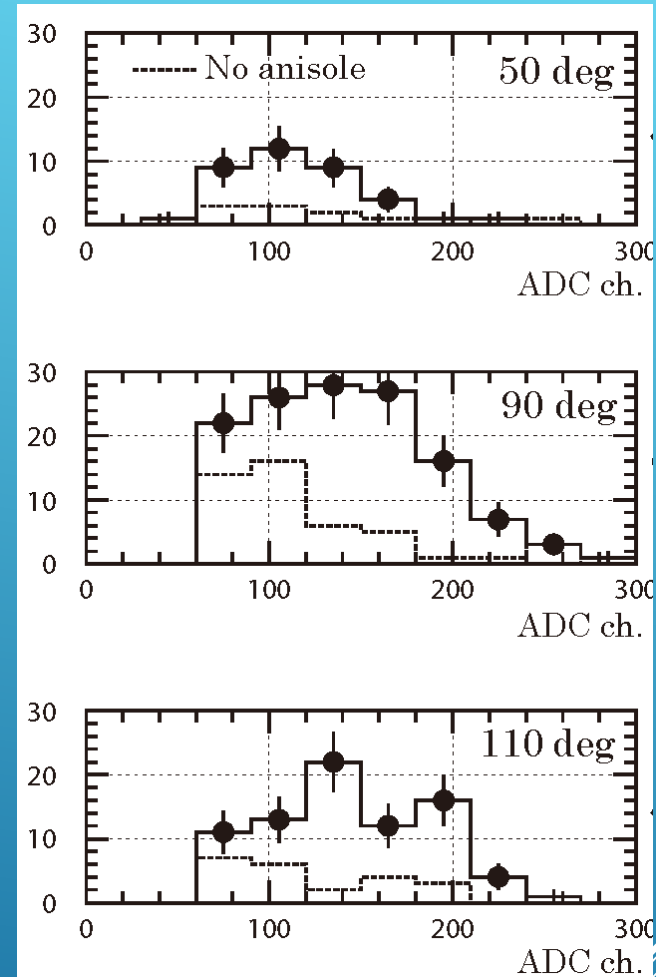
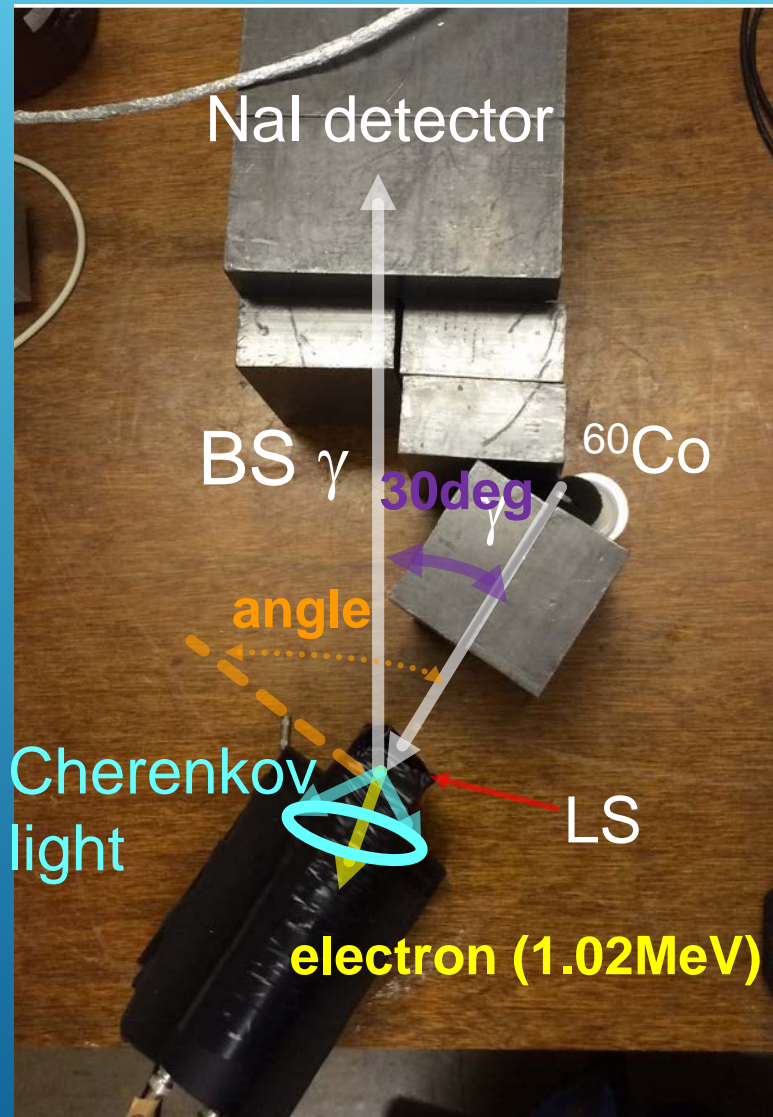
- CAEN V1721 8 channel 8bit 500MS/s FADC
- CAEN V2718 VME-PCI Optical Link Bridge

Pulse shape of scintillation light



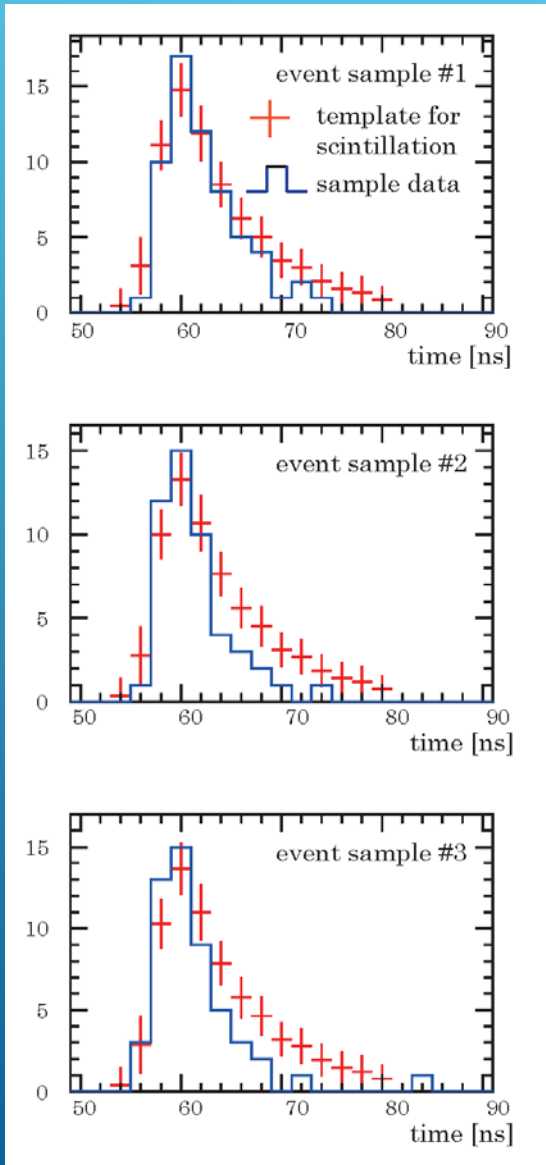
- Templates of FADC timing pulse shape for **scintillation light** were obtained for both case.
- Both decay time of scintillation light are same, and it was about 7ns.

Observation of Cherenkov lights



Cherenkov lights from $O(1\text{MeV})$ electron seem to have a directionality.

Pulse shape observed for Anisole



- Most of observed events have a different pulse shape from that of scintillation light.
- It is faster rise time and decay time.

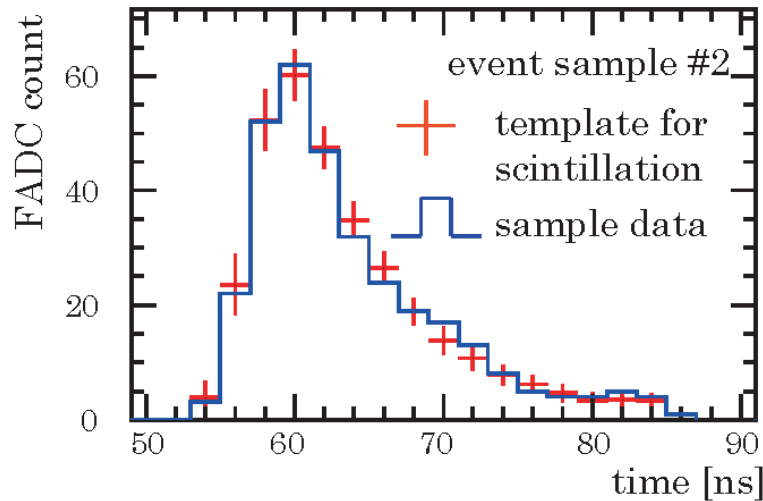


Those events mainly consist of **Cherenkov lights**, because of low QE for wave length of scintillation light (300nm).

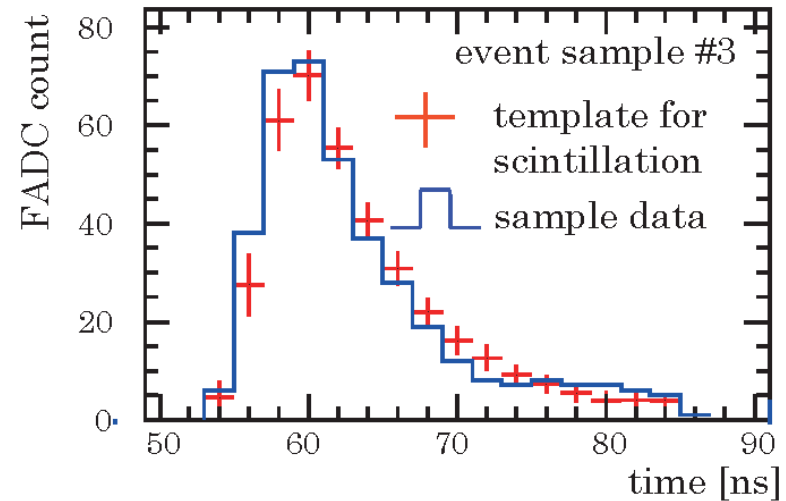
Same pulse shapes were also observed in H₂O.

Pulse shape observed for Zr loaded LS

Scintillation-only



Scinti.+Cherenkov-like

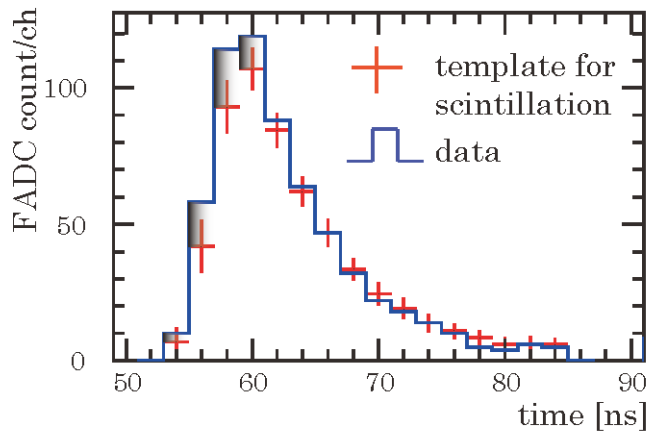


Mainly two types of pulse shape (Scintillation-only and Scinti.+Cherenkov-like) were observed.

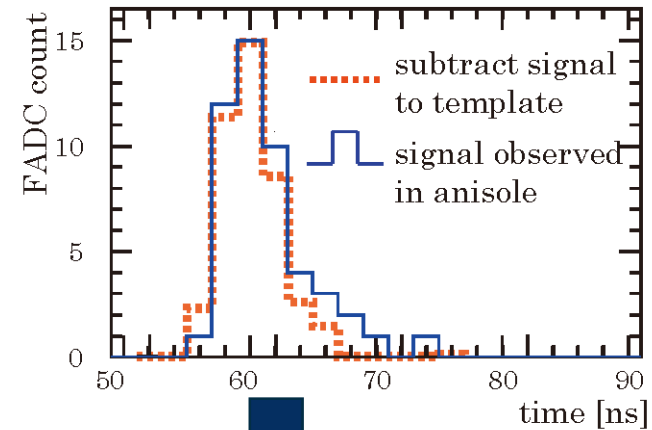
We maybe use pulse shape discrimination for the selection of events which include Cherenkov lights.

Pulse shape discrimination

Typical pulse shape of Scinti.+Cherenkov-like



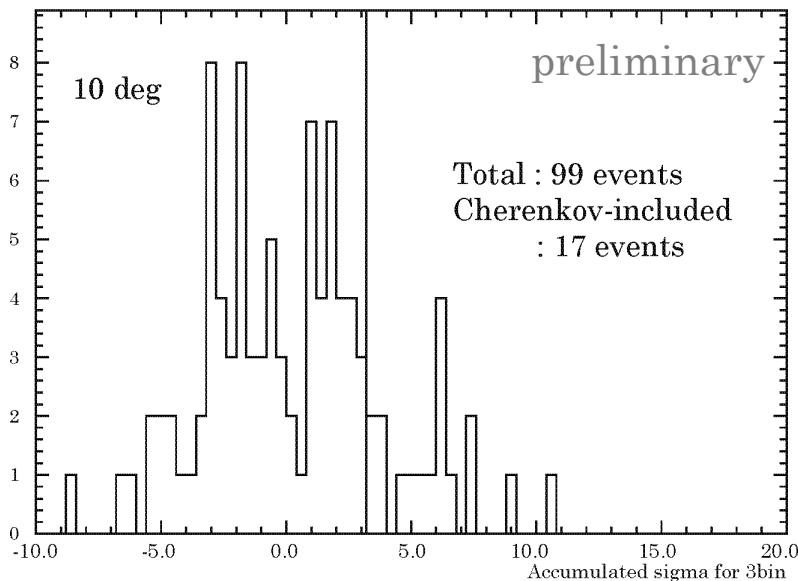
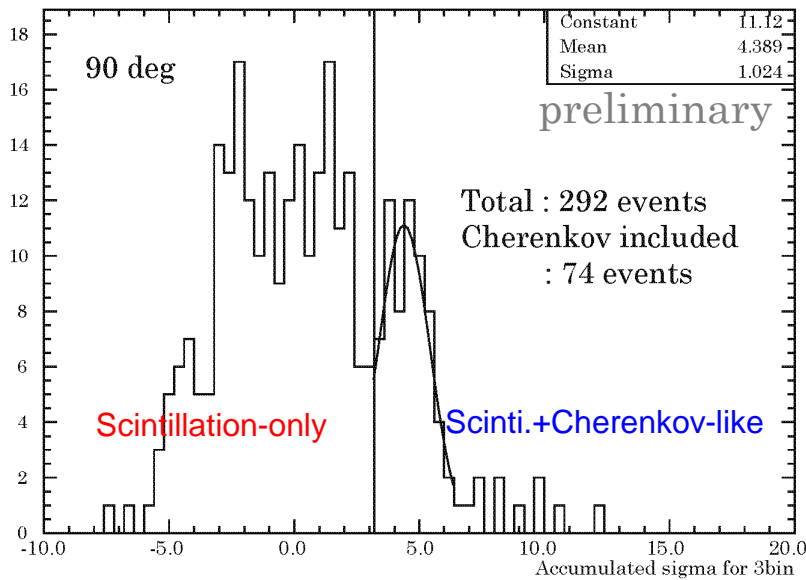
Comparison of excess pulse shape and signal for anisole



Excess pulse shape seems to be consistent with the pulse which was seen in Anisole (see p12).

Excess of sigma between data and template were accumulated for first 3 bins : accumulated sigma

Accumulated sigma distribution



Ratio of Scintillation+Cherenkov

- 90deg $25.3 \pm 3.3 \%$
 - 10deg $17.2 \pm 4.5 \%$
- Significance : 1.5σ


Scinti.+Cherenkov-like might be discriminated by acc. sig. method... but

The difference of ratio was smaller than expected by Anisole.



There seems to exist some different conditions between anisole and LS.

Summary

- ▶ Conceptual design of ZICOS detector (10 wt.% $\text{Zr}(\text{iprac})_4$ loaded Liquid Scintillator has **2.8%** **@3.35MeV** energy resolution assuming 64% photo coverage of 20" PMT) for next generation DBD experiment. ($T_{1/2}(0\nu\beta\beta) > 10^{27}$ years).
 - ^{96}Zr : **45kg** (nat.)  **865kg** (50% enrich)
 - Further **1/20 reduction of ^{208}Tl backgrounds** using PMT hit pattern of Cherenkov lights.
- ▶ PSD could be useful for the extraction of Cherenkov lights, however still need to study for confirmation.
- ▶ If PSD works, then **BG reduction using PSD** should be checked by prototype for next step.

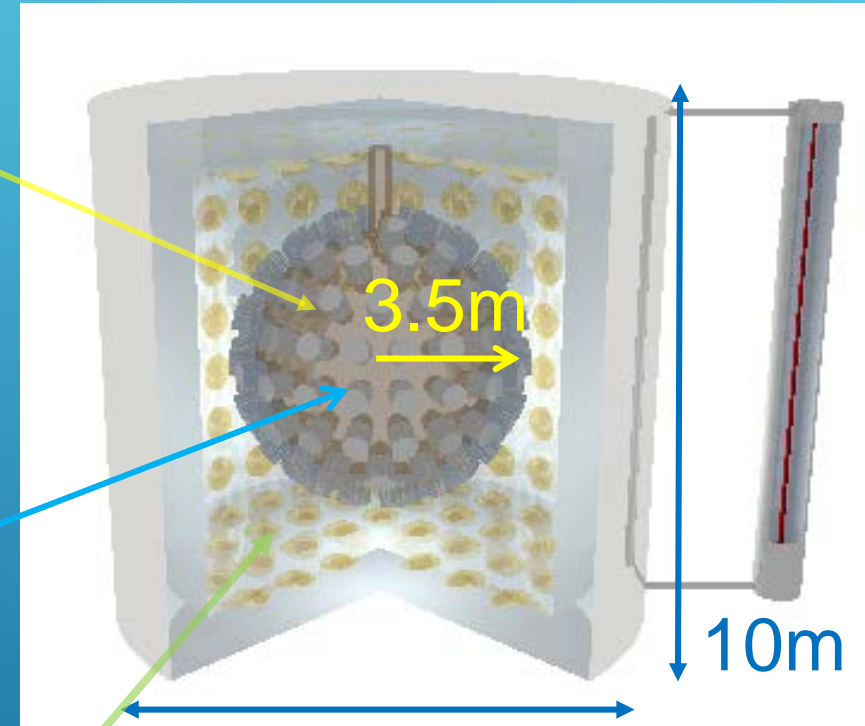
BACKUP

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

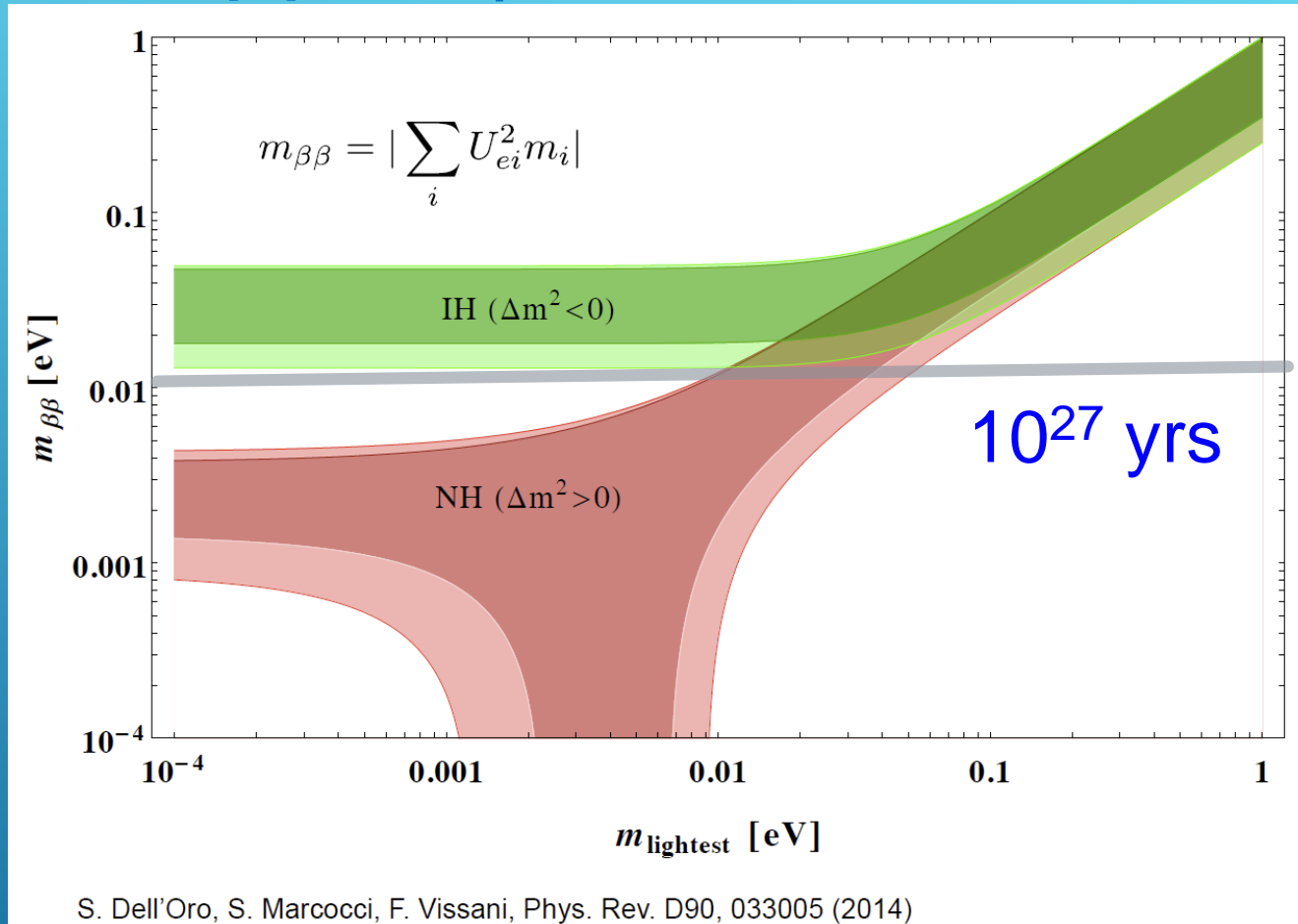
1.5wt.% Zr loaded Liquid Scintillator :
Light yield of $48.7 \pm 7.1\%$ for BC505,
and an energy resolution of $2.8 \pm 0.4\%$
at 3.35 MeV assuming 64% photo
coverage of the photomultiplier

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

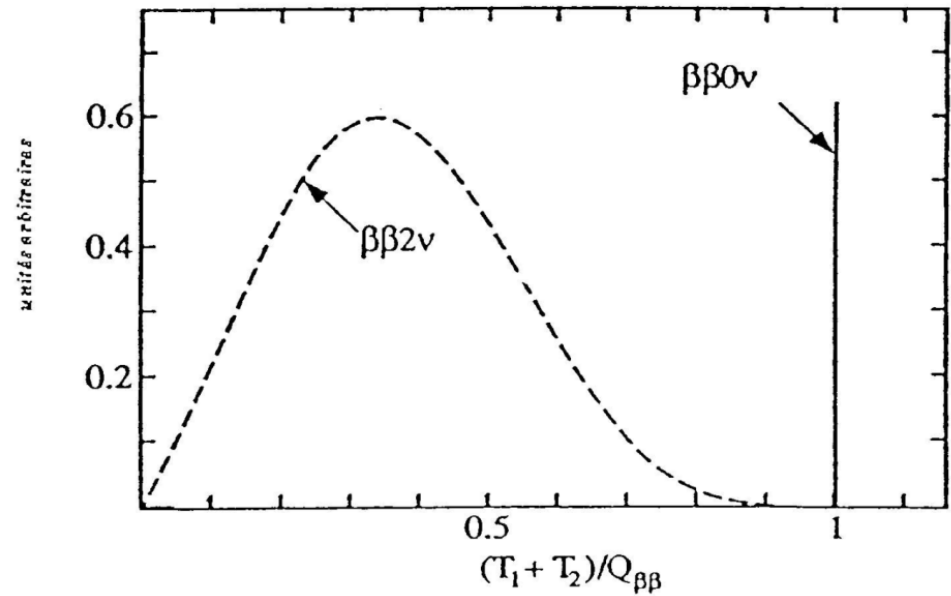


~tons of target and ~zero BG detector will be necessary for next generation $0\nu\beta\beta$ experiment.

Neutrinoless double beta decay

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

Transition	$Q_{\beta\beta}$ (keV)	Abundance (%) ($^{232}\text{Th} = 100$)
$^{110}\text{Pd} \rightarrow ^{110}\text{Cd}$	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



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

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

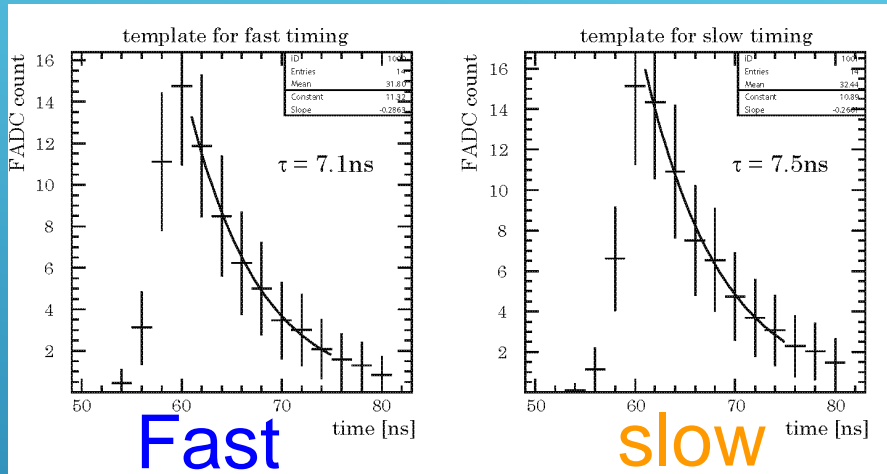
a: abundance M: target mass

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

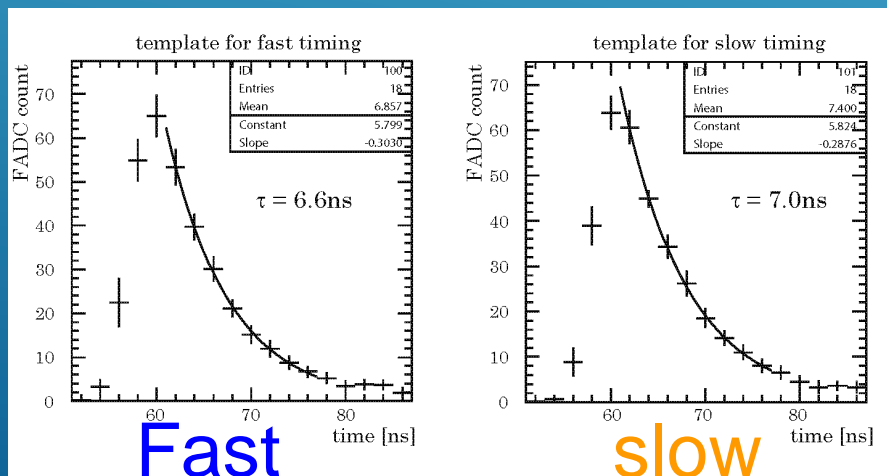
Requirement : Low BG, Large target mass, High E-resolution

Pulse shape of timing information

Anisole only



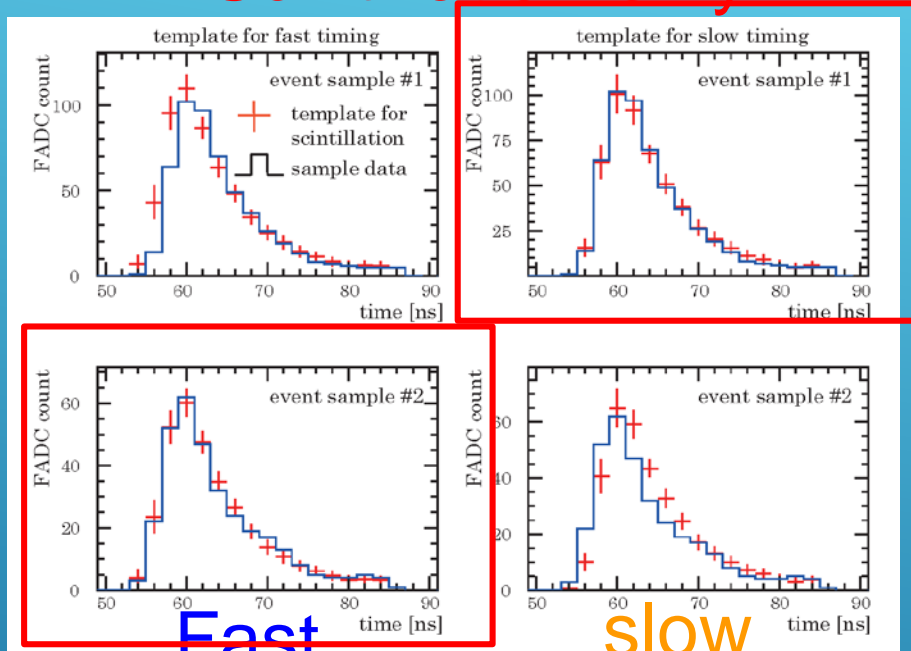
Zr(iprac)₄ loaded LS



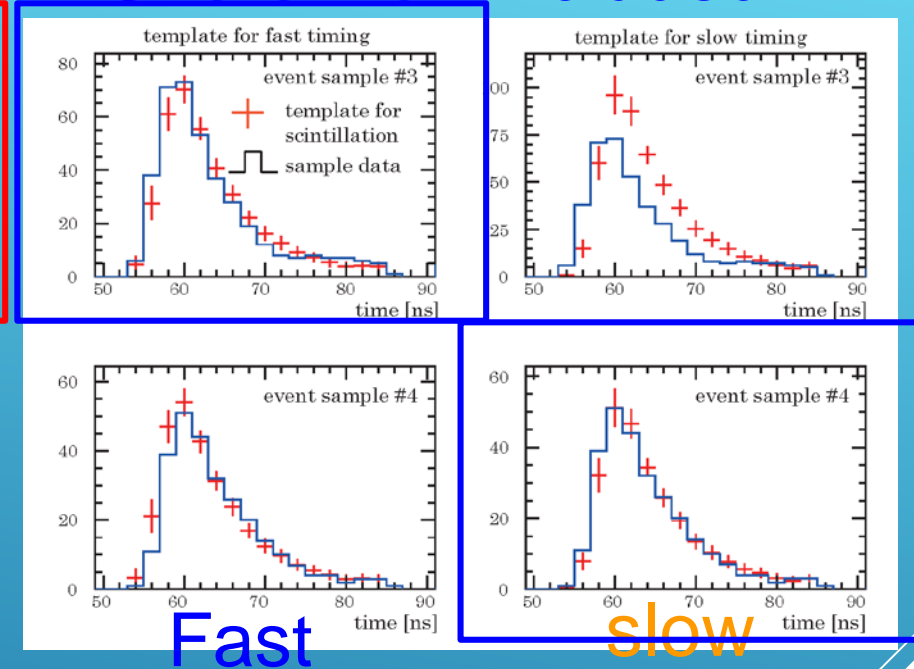
- Templates of pulse shape of timing information for **scintillation light** were obtained by FADC.
- Fast and slow rise time component were observed due to FADC resolution.
- Both decay time of scintillation light are about 7ns.

Pulse shape observed in Zr loaded LS

Scintillation-only



Cherenkov-included

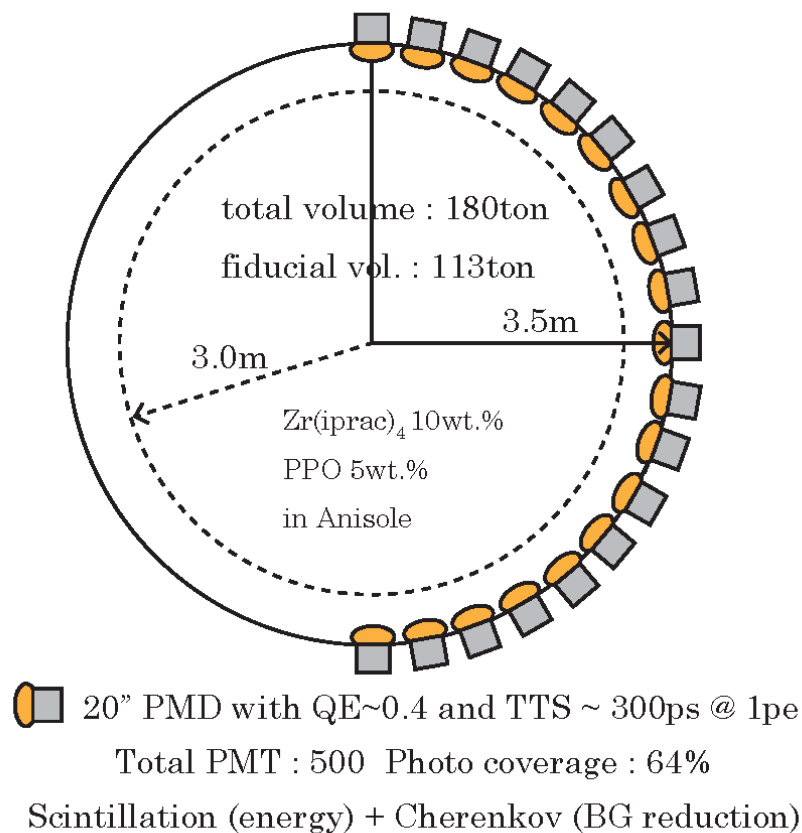


Two types of pulse shape (Scintillation-only and Cherenkov-included) were observed.

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

Design of ZICOS detector

Conceptual design of ZICOS detector



Detector :

- 1) 180tons LS : 1.5 wt.% Zr and 5wt.% PPO in Anisole .
- 2) Need 500 of 20" PMT with high QE ~0.4 and TTS ~300ps@1pe for 64% photo coverage.

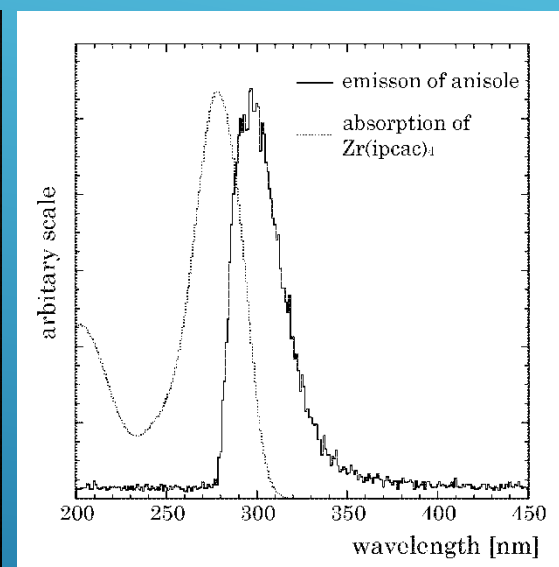
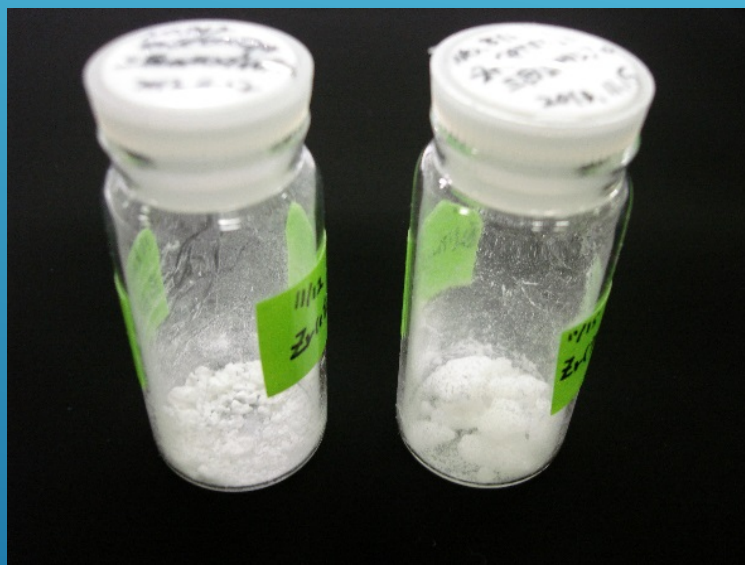
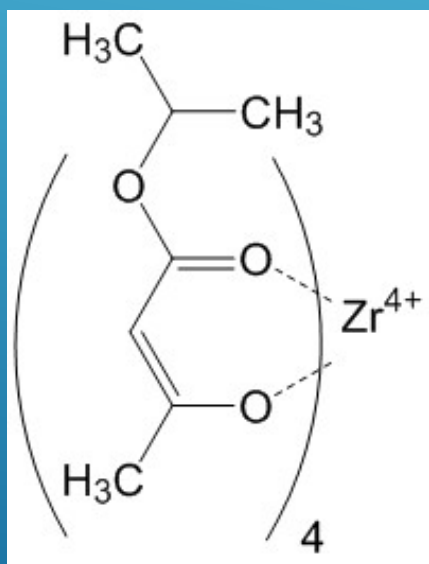
Expected performance :

- 1) Energy resolution ~2.8% @ 3.35MeV
- 2) $T_{1/2}(0\nu\beta\beta) > 10^{27}$ years, if both 1/20 BG reduction and 50% ⁹⁶Zr enrichment could be achieved.

Natural abundance of ⁹⁶Zr : 2.6%

tetrakis(isopropyl acetoacetate) zirconium

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



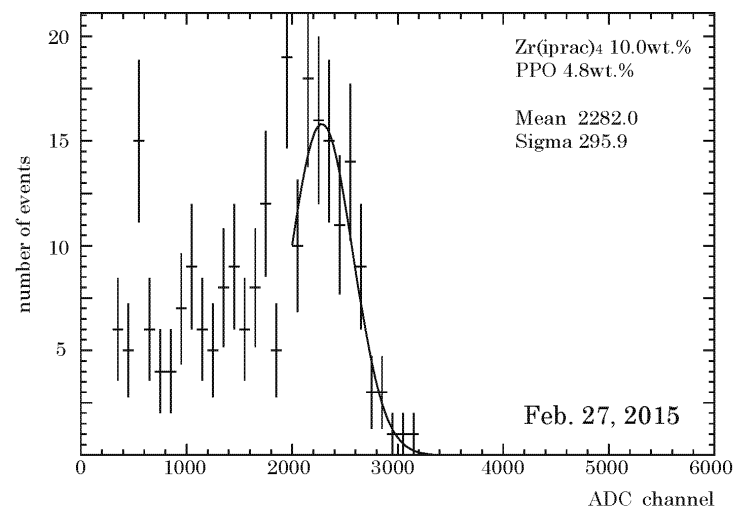
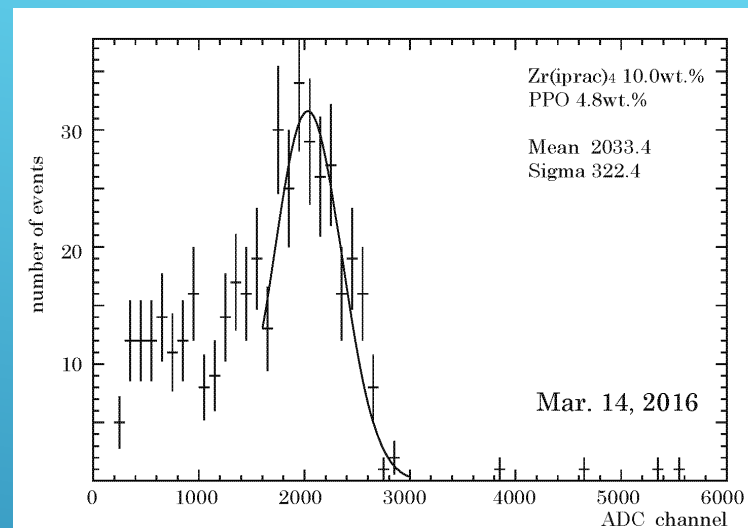
**LS = $\text{Zr}(\text{iprac})_4$: 10 wt.% PPO: 5wt.%
(POPOP: 0.05wt.%)**

Zr loaded liquid scintillator

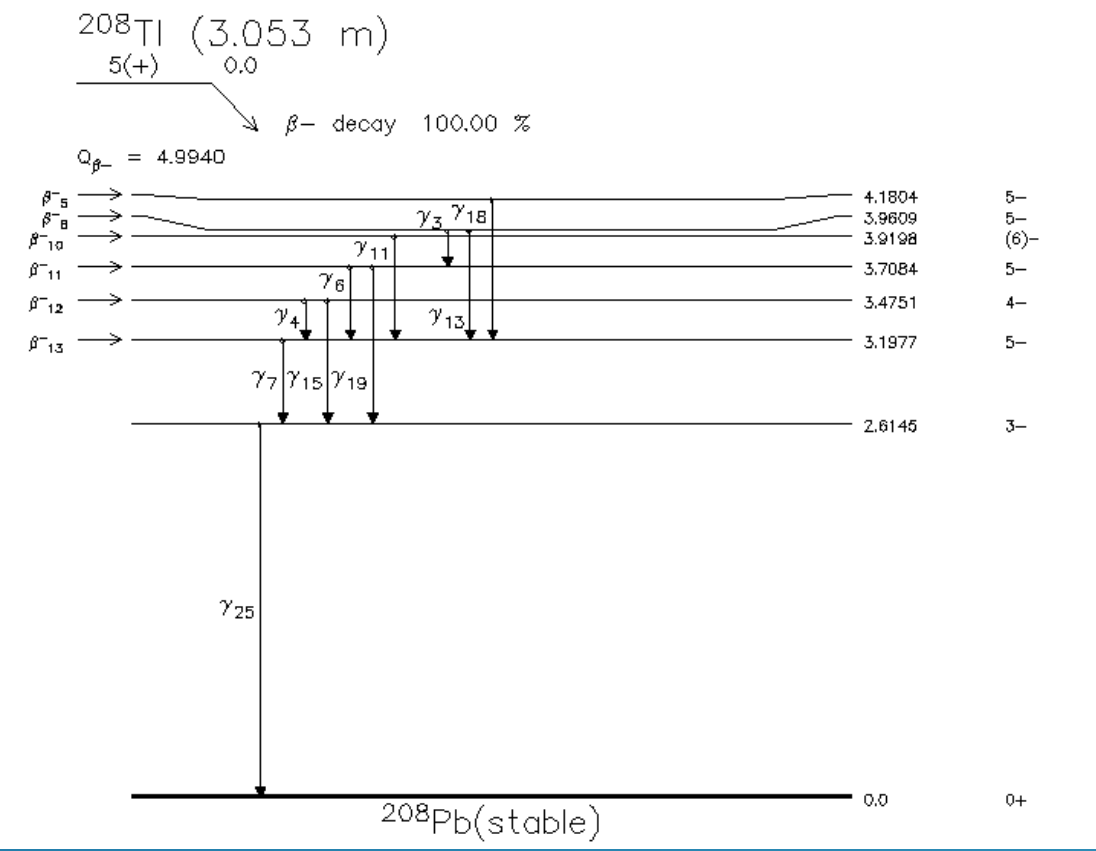
Feb. 27, 2015 Mar. 14, 2016



Light yield of $48.7 \pm 7.1\%$ for BC505, and an energy resolution of $4.1 \pm 0.6\%$ at 3.35 MeV assuming 40% photo coverage of the photomultiplier



Decay branch of Thallium-208

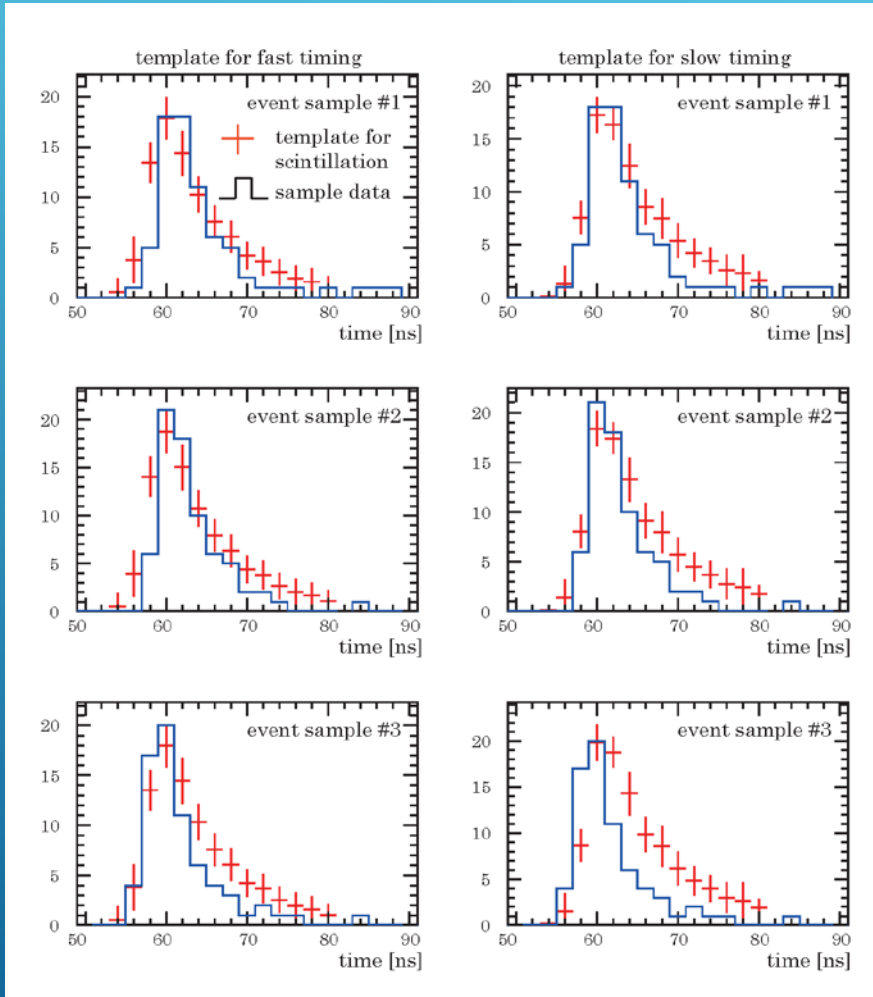


Radiations	$y(i)$ (Bq-s) ⁻¹
beta- 5	2.27×10^{-03}
beta- 8	3.09×10^{-02}
beta- 10	6.30×10^{-03}
beta- 11	2.45×10^{-01}
beta- 12	2.18×10^{-01}
beta- 13	4.87×10^{-01}
ce-K, gamma 3	4.04×10^{-03}
gamma 4	6.31×10^{-02}
ce-K, gamma 4	2.84×10^{-02}
ce-L, gamma 4	4.87×10^{-03}
gamma 6	2.26×10^{-01}
ce-K, gamma 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}

The vertex reconstructed by scintillation make it within fiducial volume due to misfitting of gammas.

Pulse shape observed in H₂O

90deg

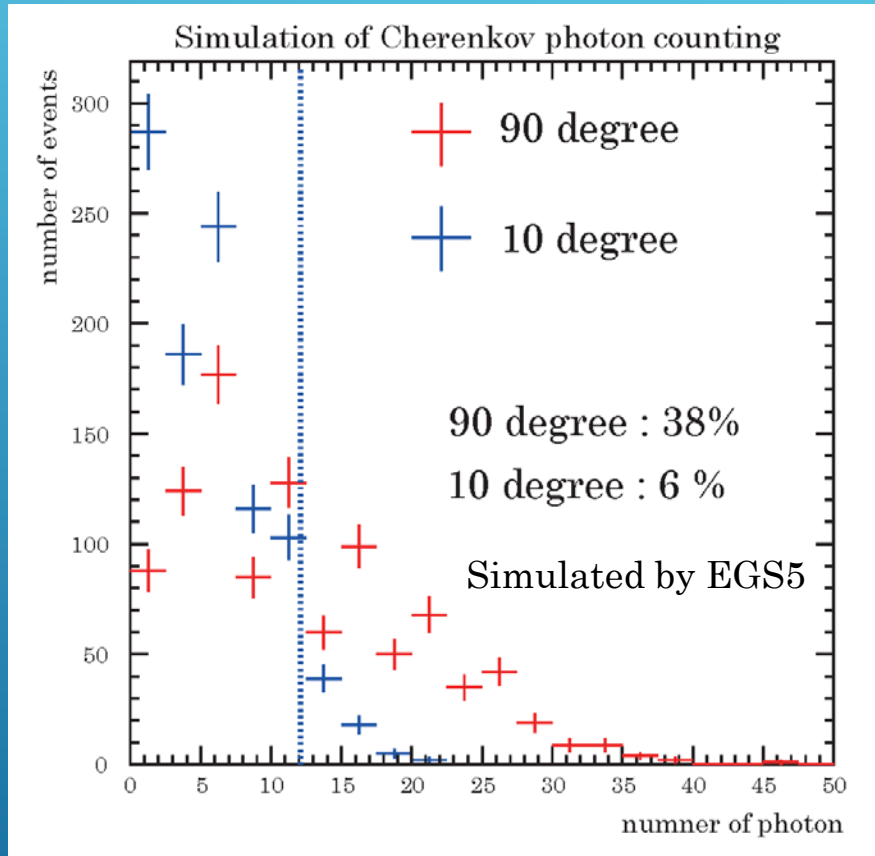


- Same pulse shape of timing as anisole was observed.
- This signal should be caused by Cherenkov light, because of no scintillation in H₂O.



This pulse shape is made by Cherenkov lights.

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.

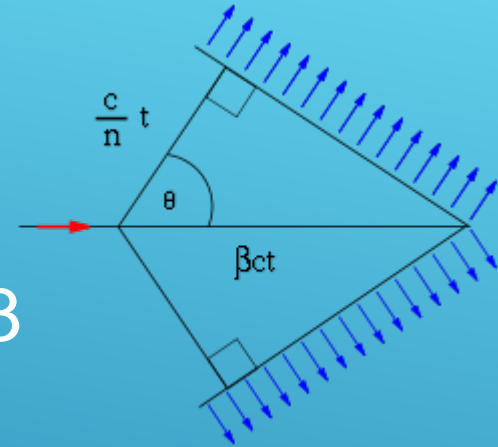


Maybe different situation in Liquid Scintillator.

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

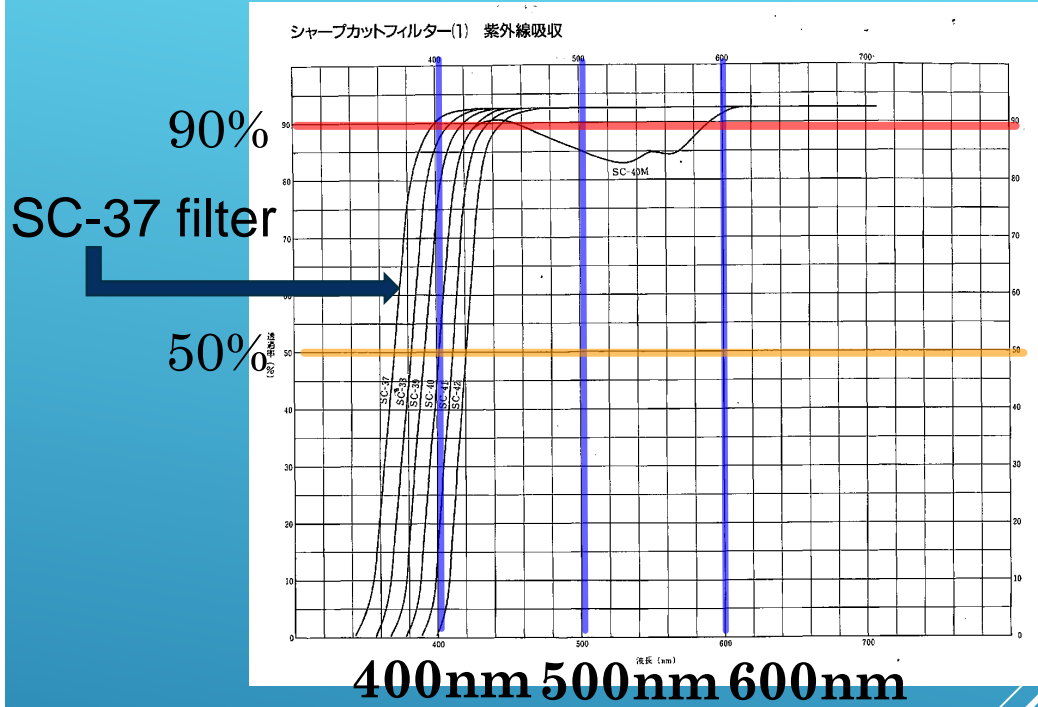
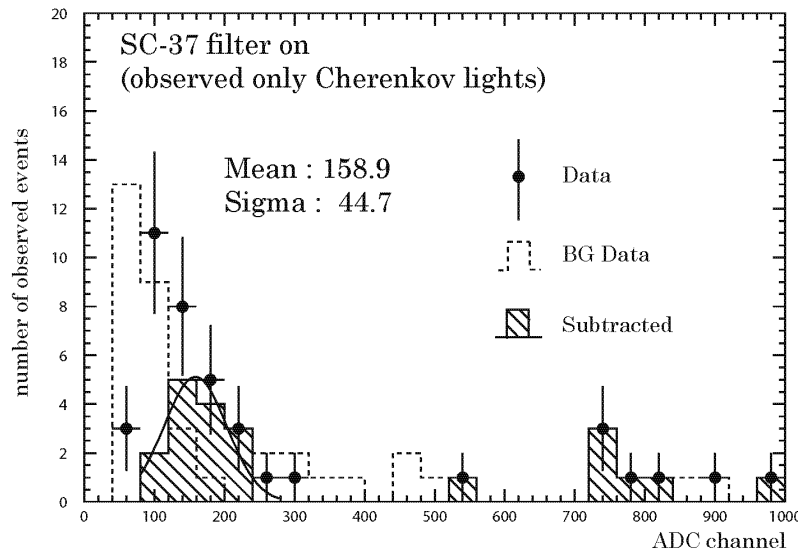
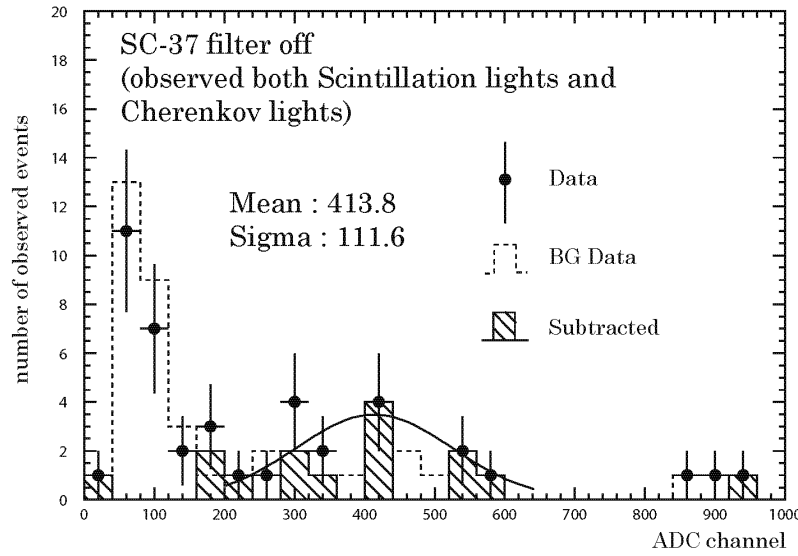


$$\frac{dN}{dx} = 2\pi z^2 \alpha \sin^2 \theta_c \int_{\lambda_1}^{\lambda_2} \frac{d\lambda}{\lambda} = 475 z^2 \sin^2 \theta_c \text{ photon/cm}$$

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

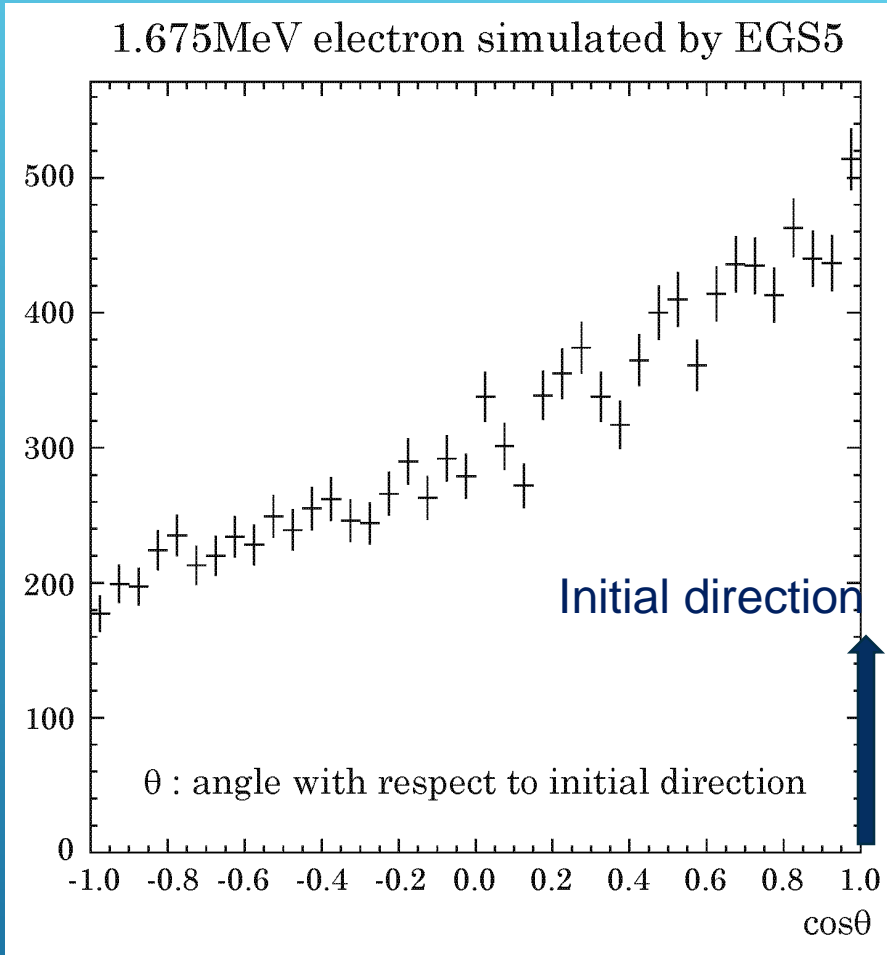
Cherenkov light = $\sim 1\%$ of scintillation light

Light yield of Cherenkov lights



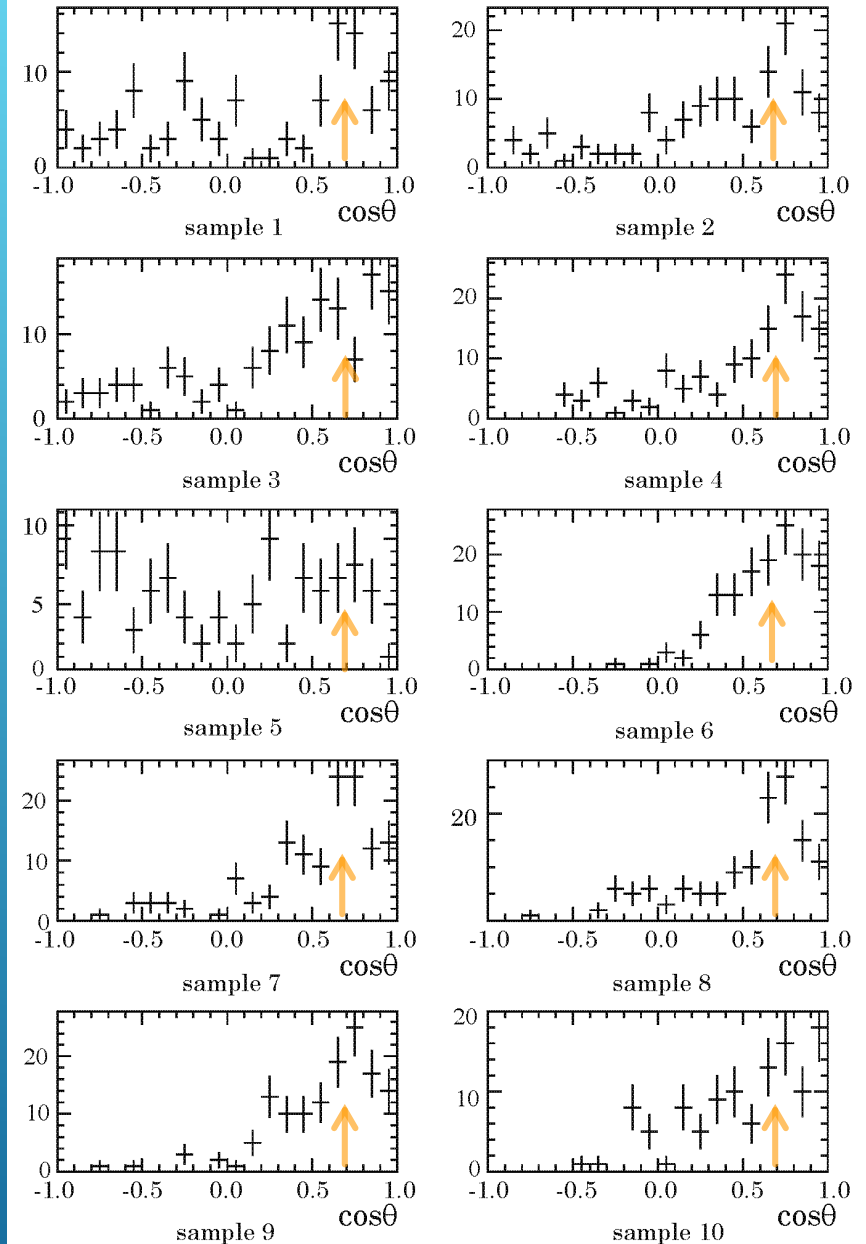
Cherenkov light yield ($\lambda > 400\text{nm}$)
Scintillation light yield of anisole
 = $\sim 0.02 \equiv \sim 200 \text{ photon/MeV}$

Multiple scattering

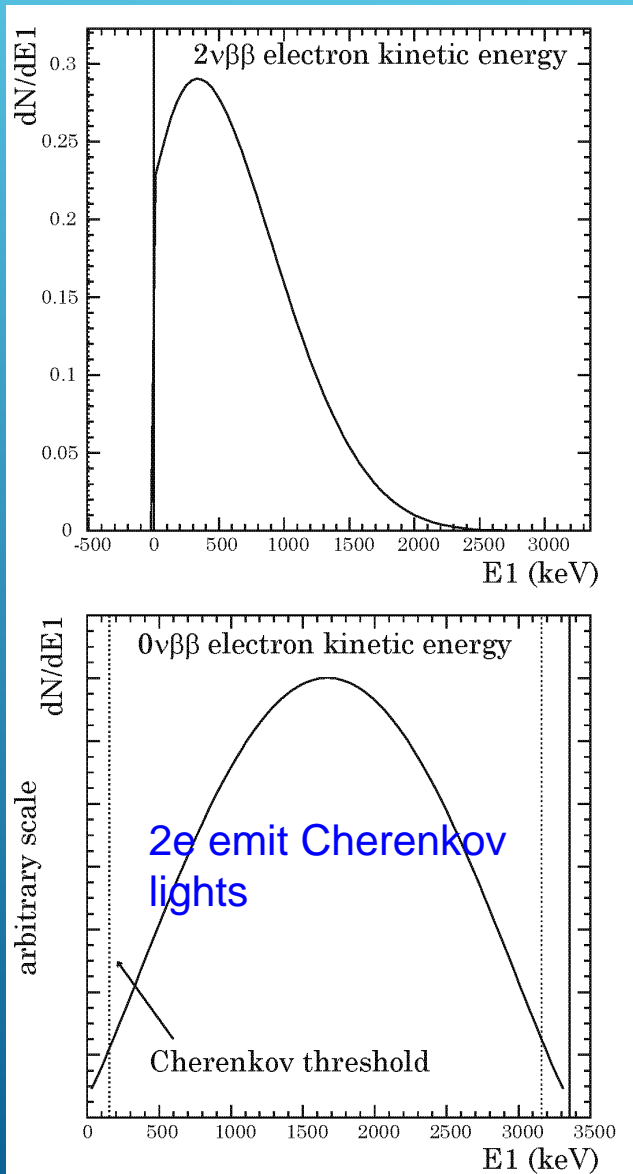


Even though multiple scattering of electrons, Cherenkov photons look have some clusters.

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

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

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

Q : Q value m_e : electron mass

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

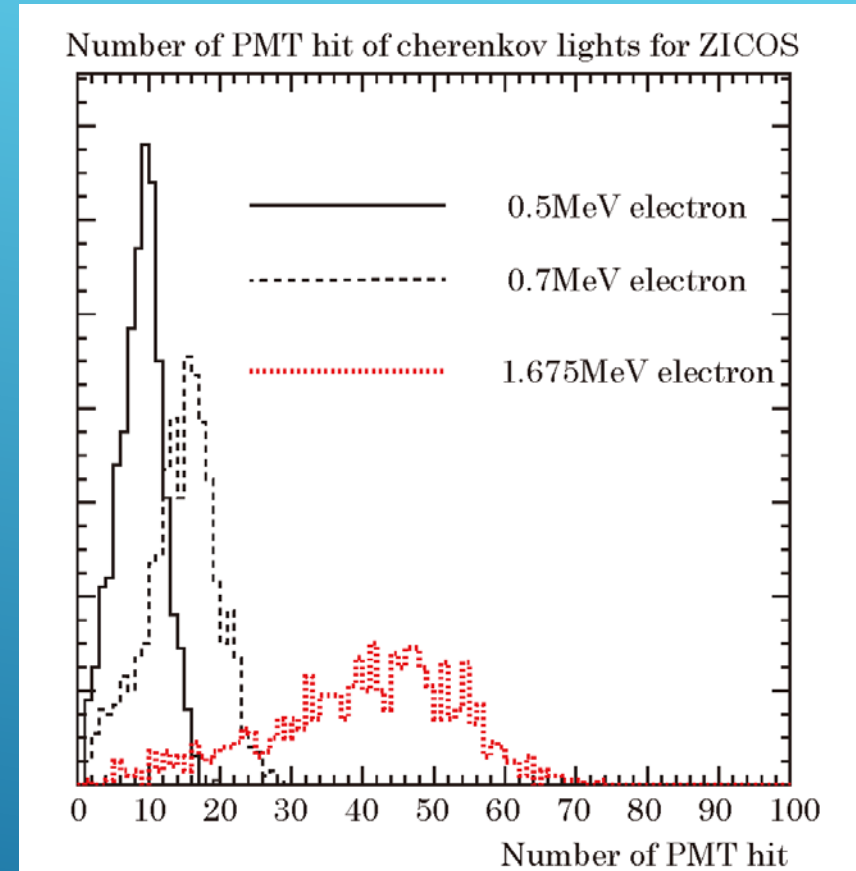
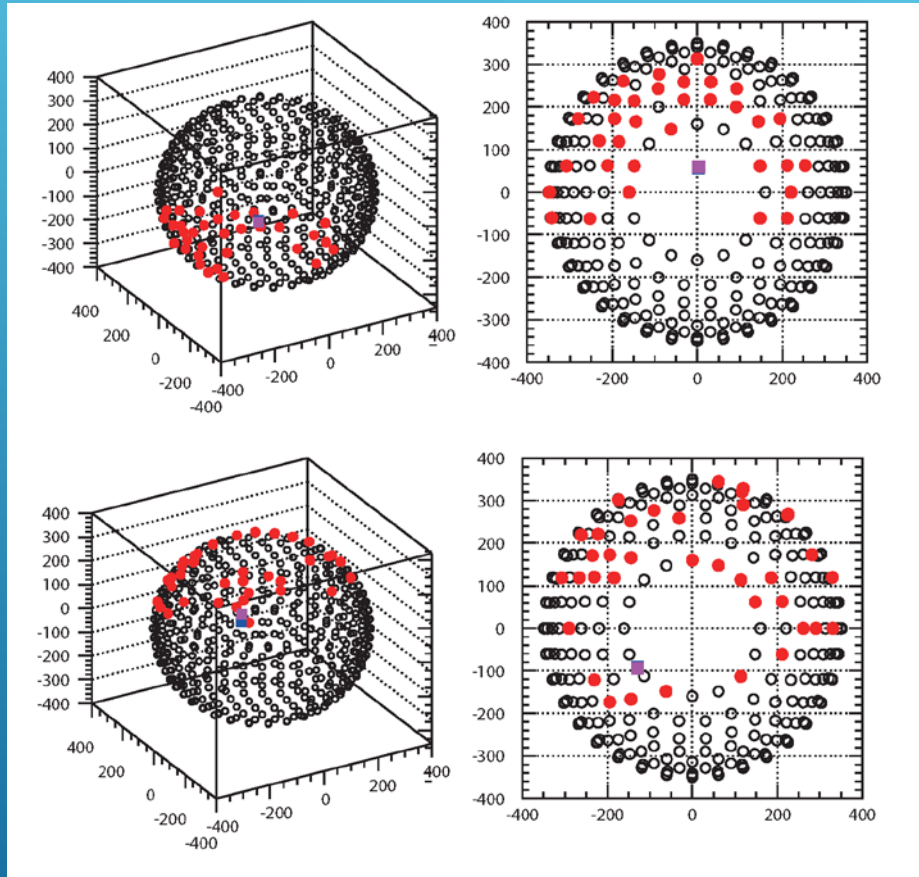
ε_i can generate independently within energy conservation.

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

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

Simulation of Cherenkov lights

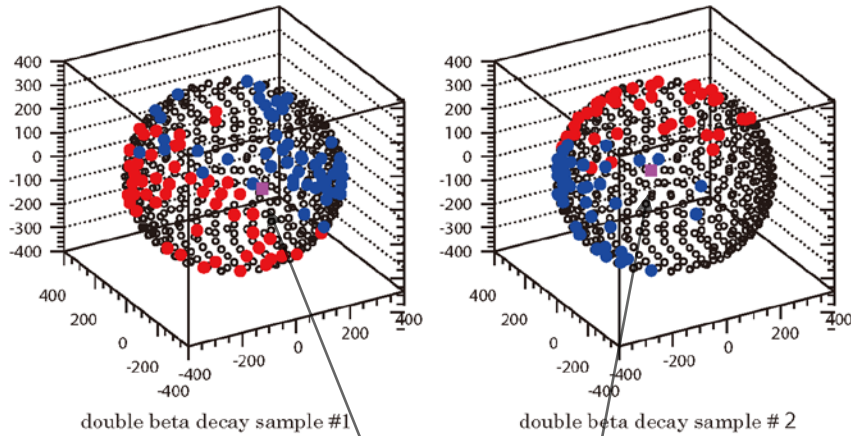
Simulated by EGS5 (kinetic energy 1.675MeV)



Hard to detect Cherenkov events below 0.5MeV.

Hit pattern of DBD (opposite and half E)

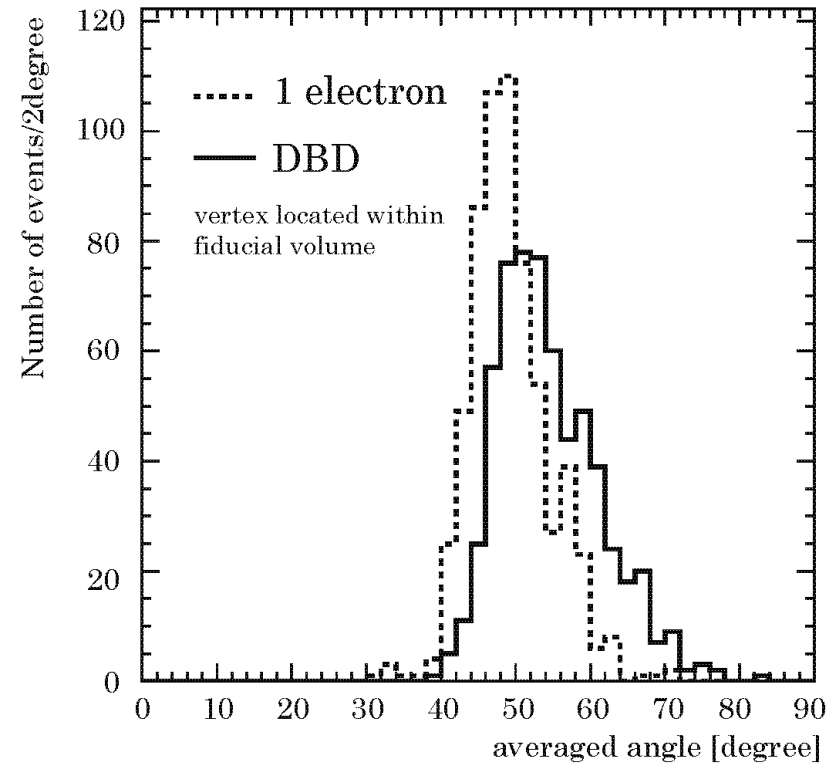
Simulated by EGS5 (kinetic energy 1.675MeV)



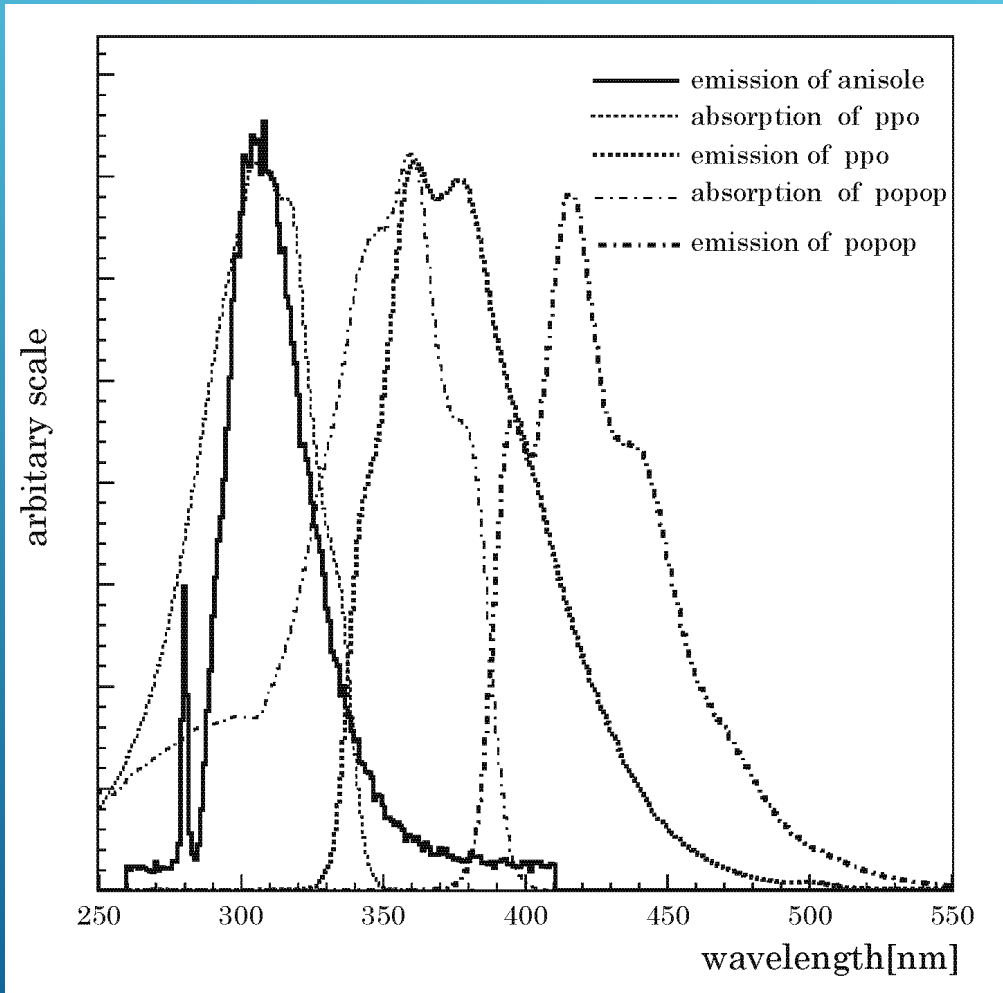
Generate position of DBD

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

averaged angle with respect to averaged direction



Emission and absorption spectra for solvent and solute in standard cocktail

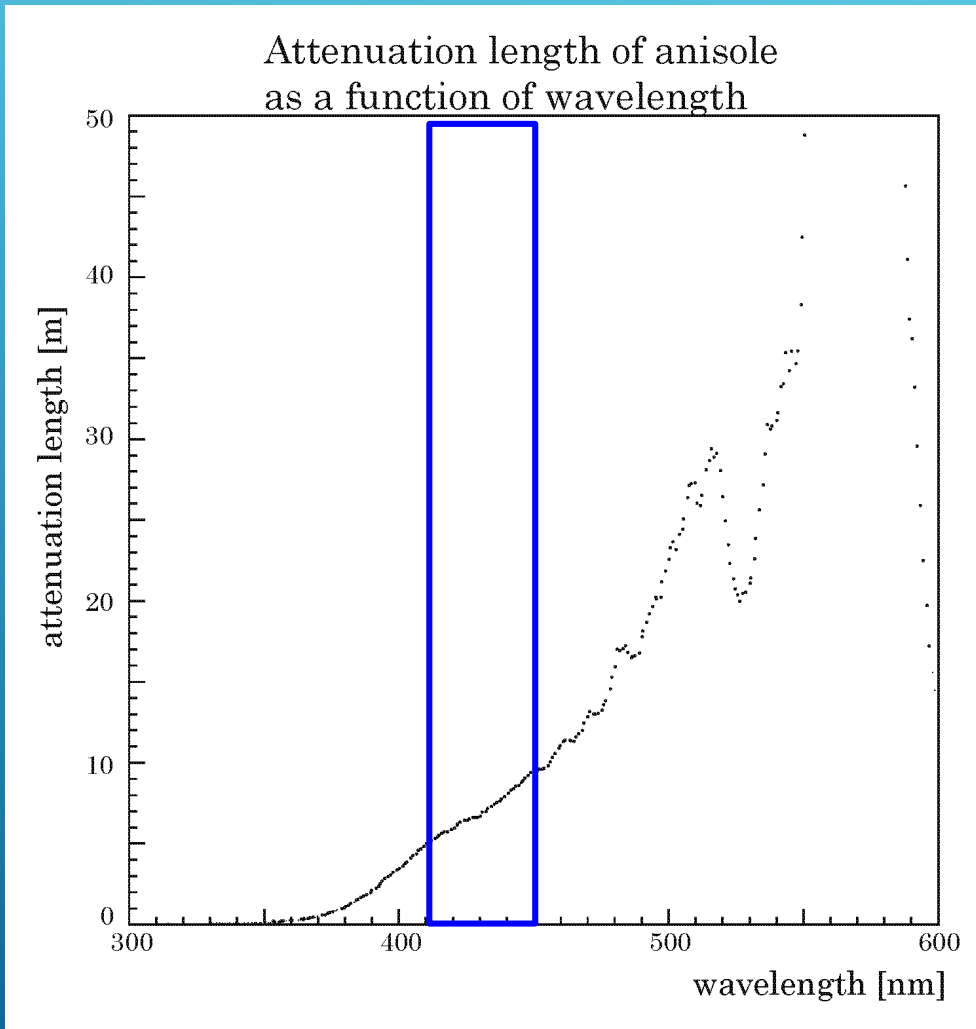


PPO absorbed most of emission lights from anisole.



Effectively the energy was transferred to the secondary scintillator.

ATTENUATION LENGTH OF ANISOLE

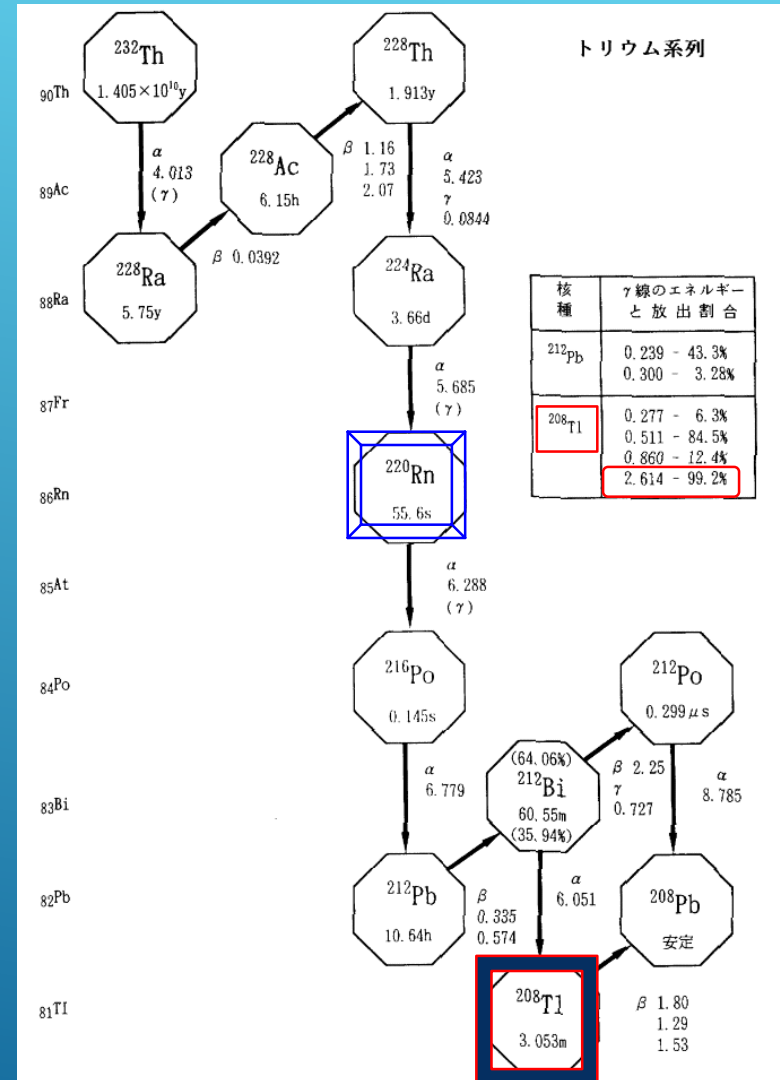
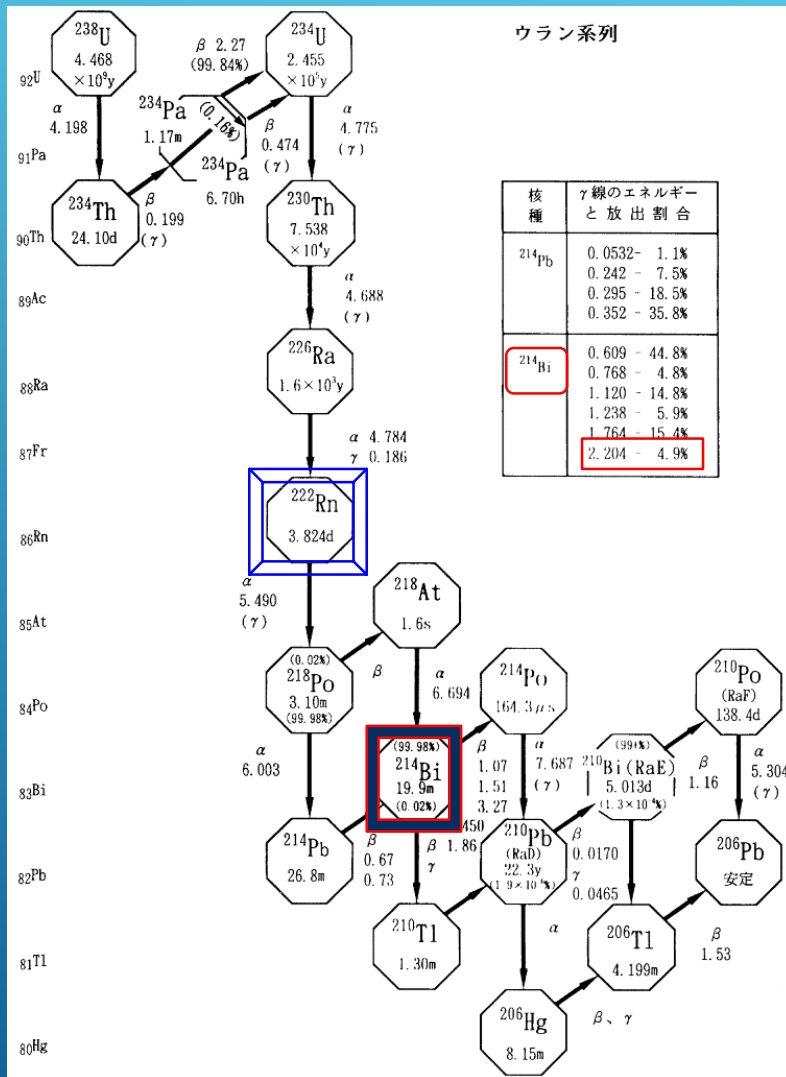


Attenuation length of scintillation light from POPOP (~450nm) was obtained as ~6m.

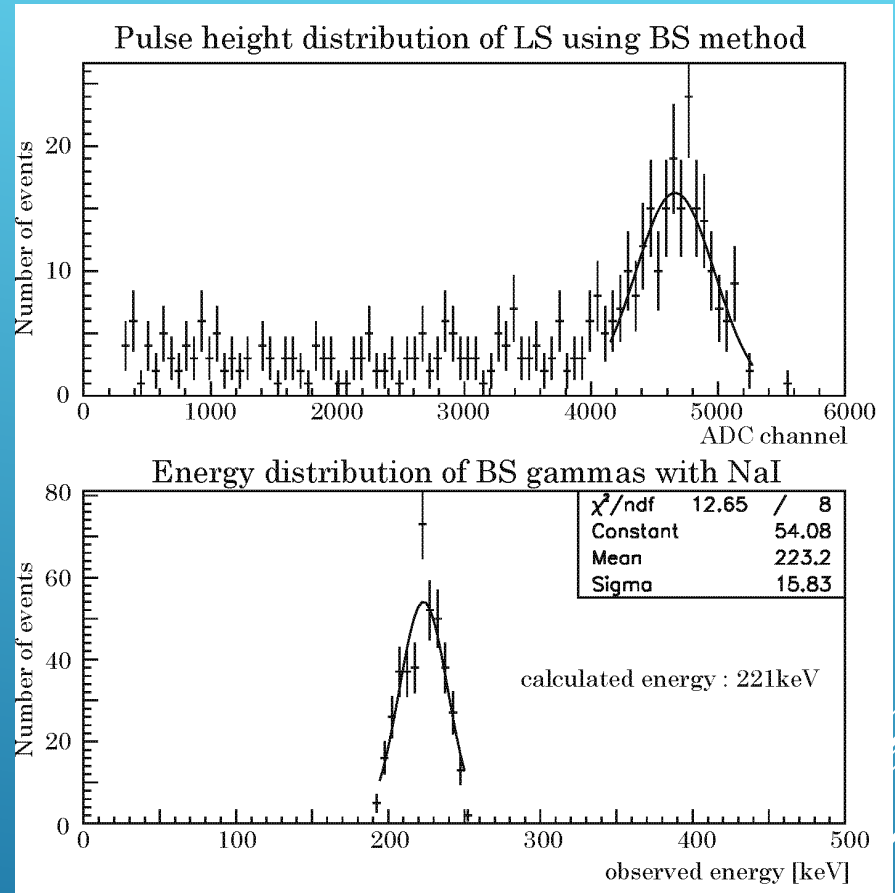
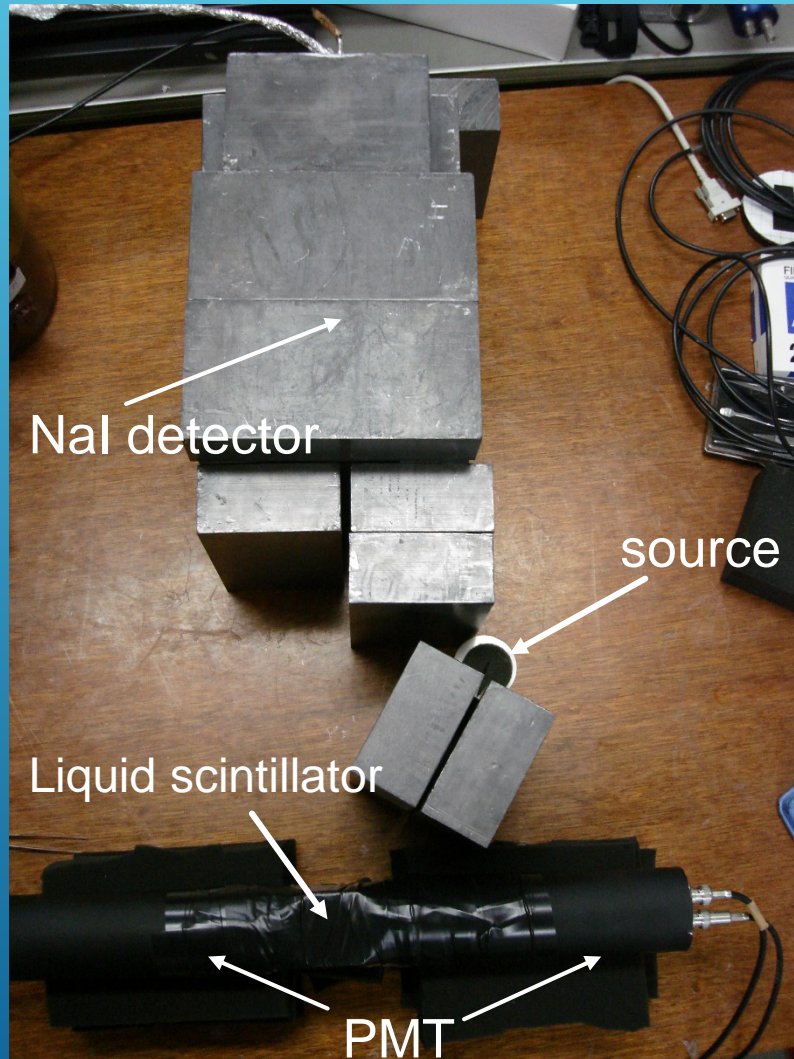


No problem for radius of ZICOS detector.

Natural radiative U/Th decay chain

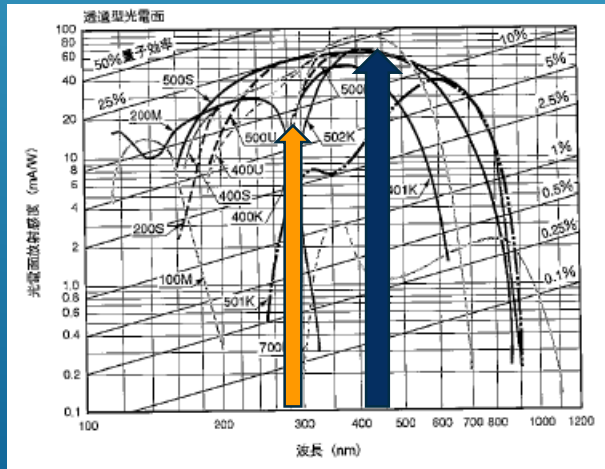
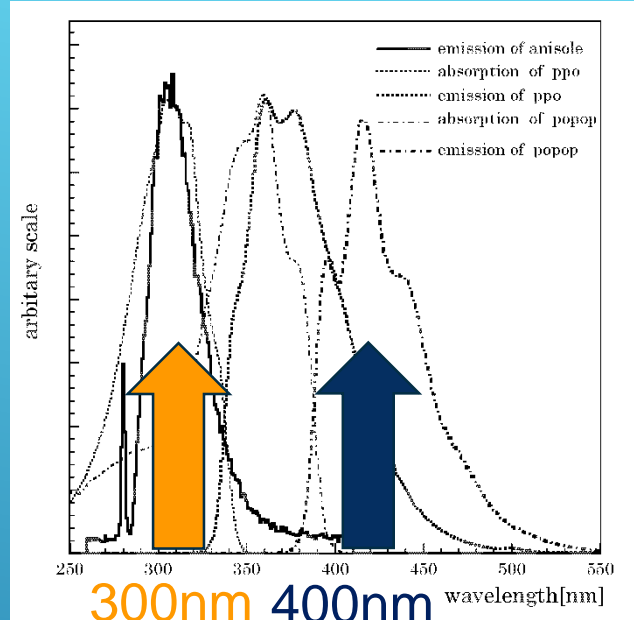
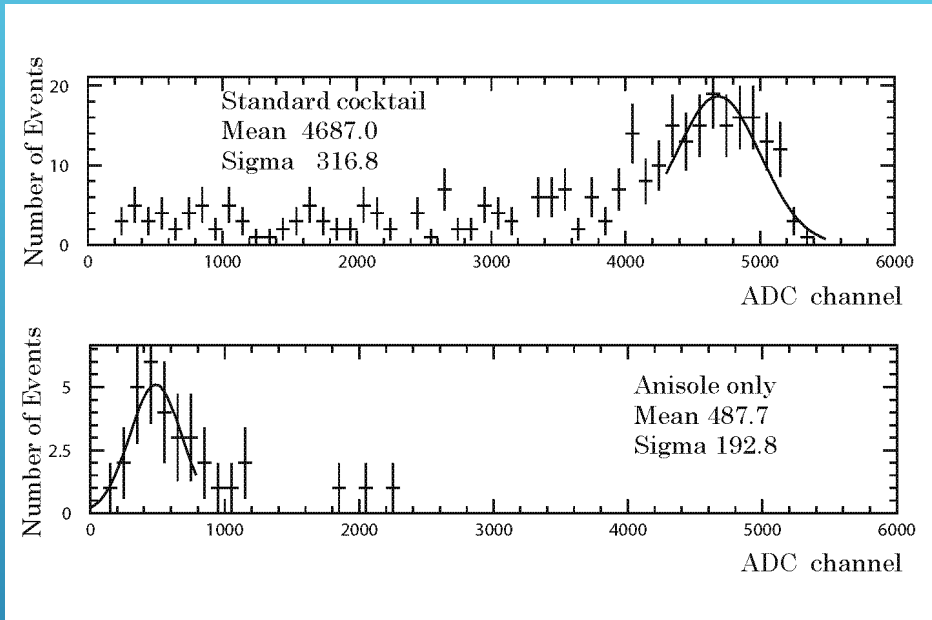


Backscattering method



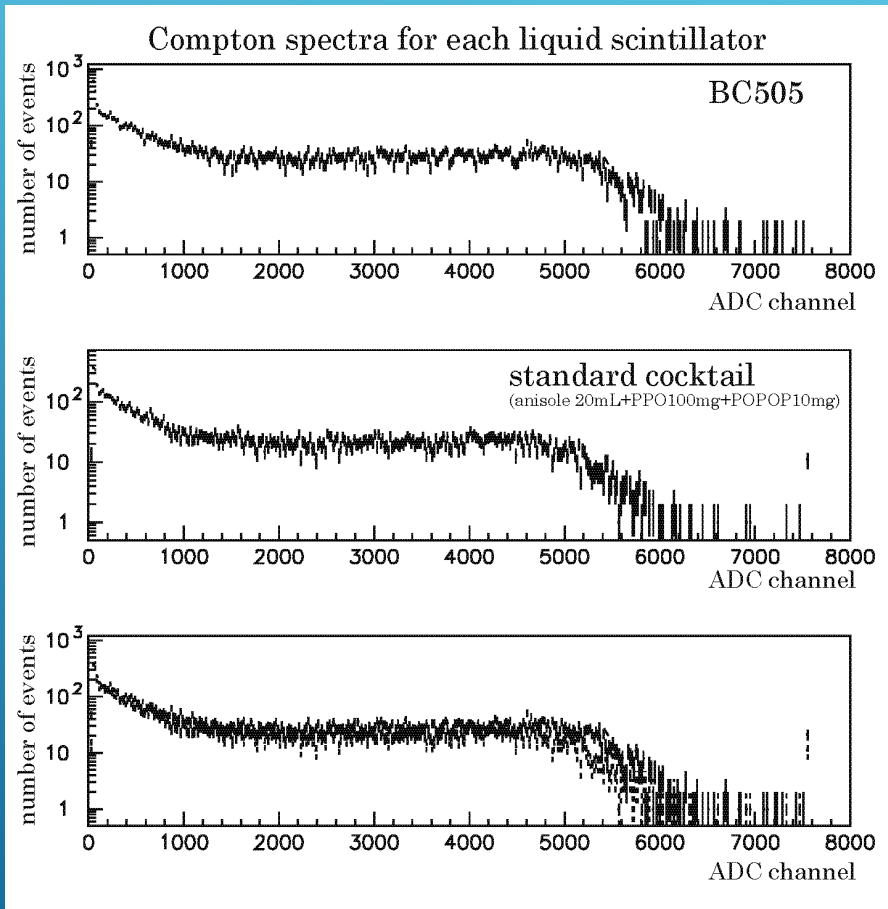
Single peak could be used even in liquid scintillator.

Light yield of scintillation in anisole



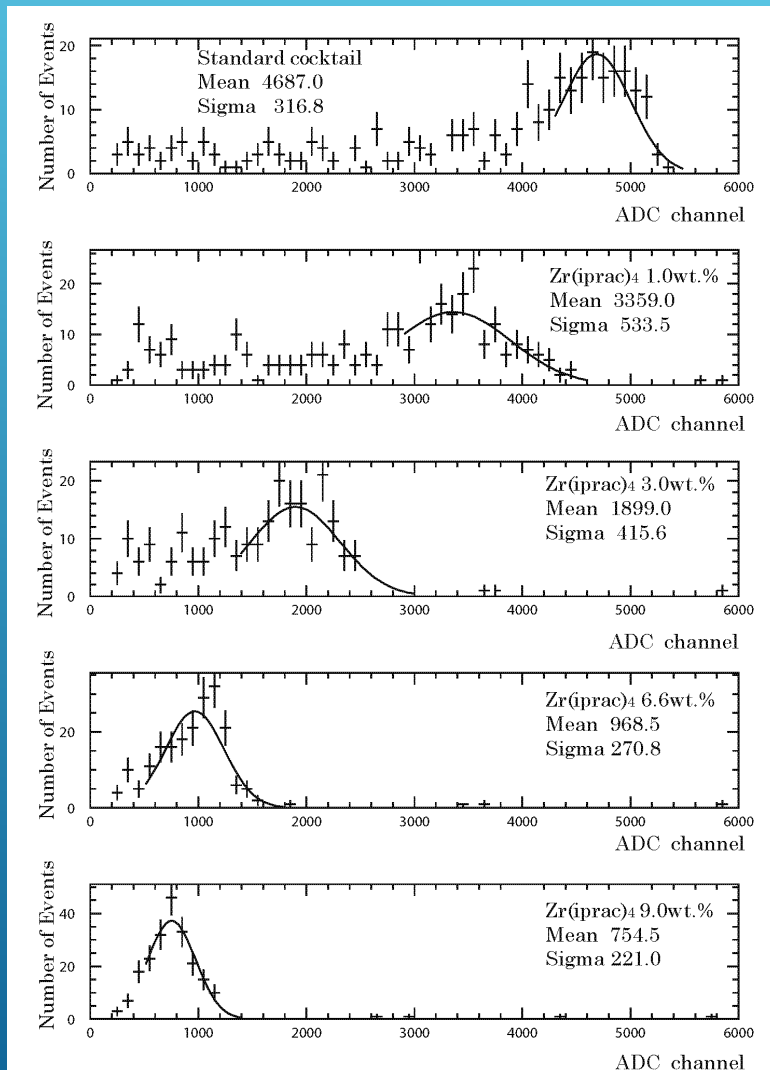
Relative scintillation light yield of **anisole** is 9.8% to **standard cocktail** (due to difference of quantum efficiency of PMT)

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

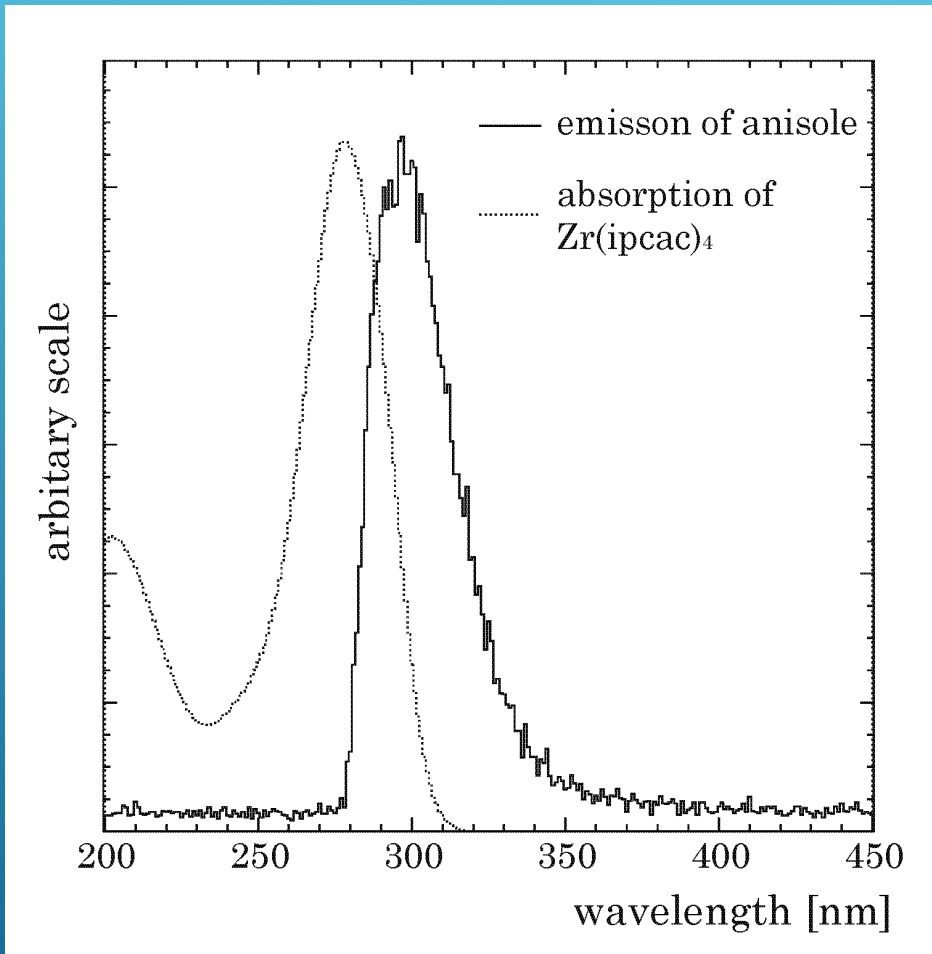
Physical Constants of SGC Liquid Scintillators

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 $Zr(iprac)_4$

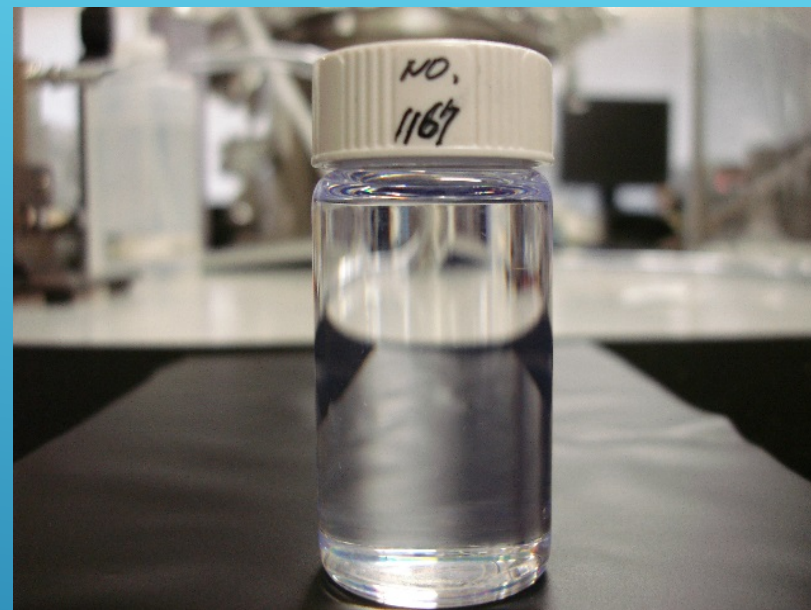
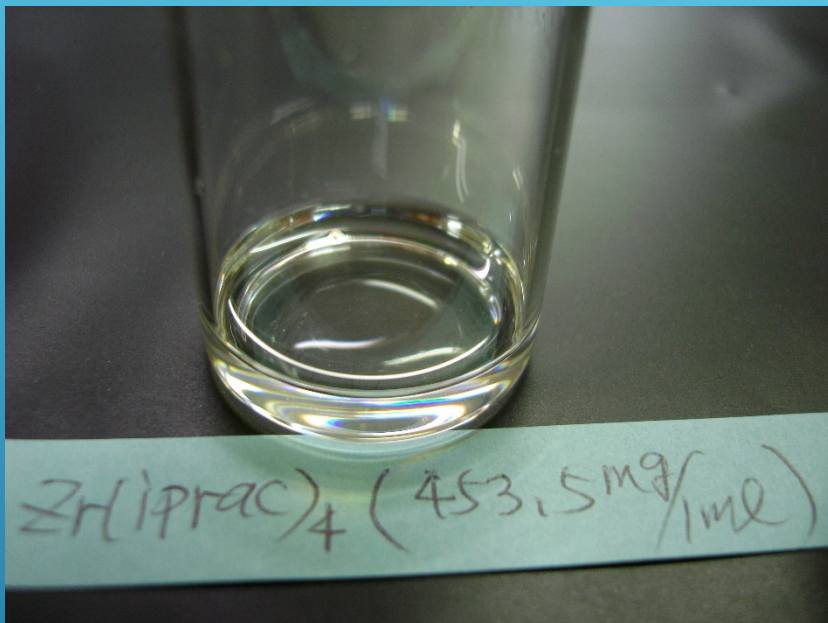


Absorption peaks of $Zr(iprac)_4$ was found around at 278nm. However, overlapped region with emission of anisole was existed.



$Zr(iprac)_4$ 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

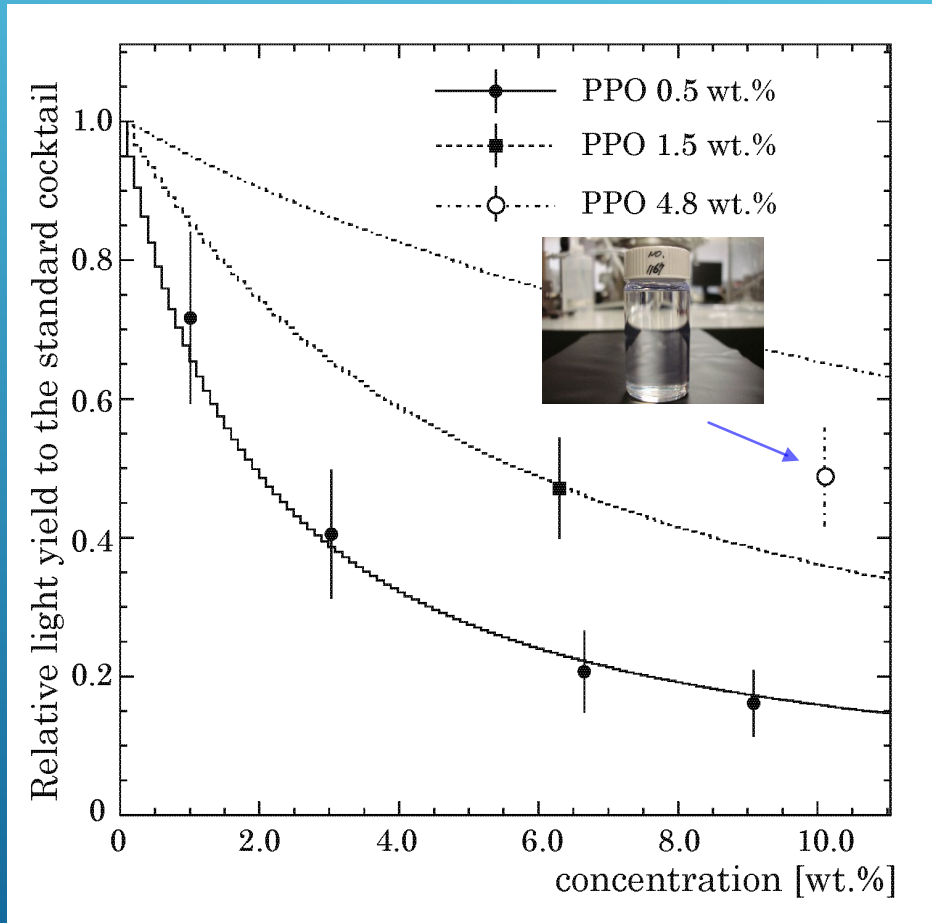
σ_1 : absorbance of PPO (mol^{-1})

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



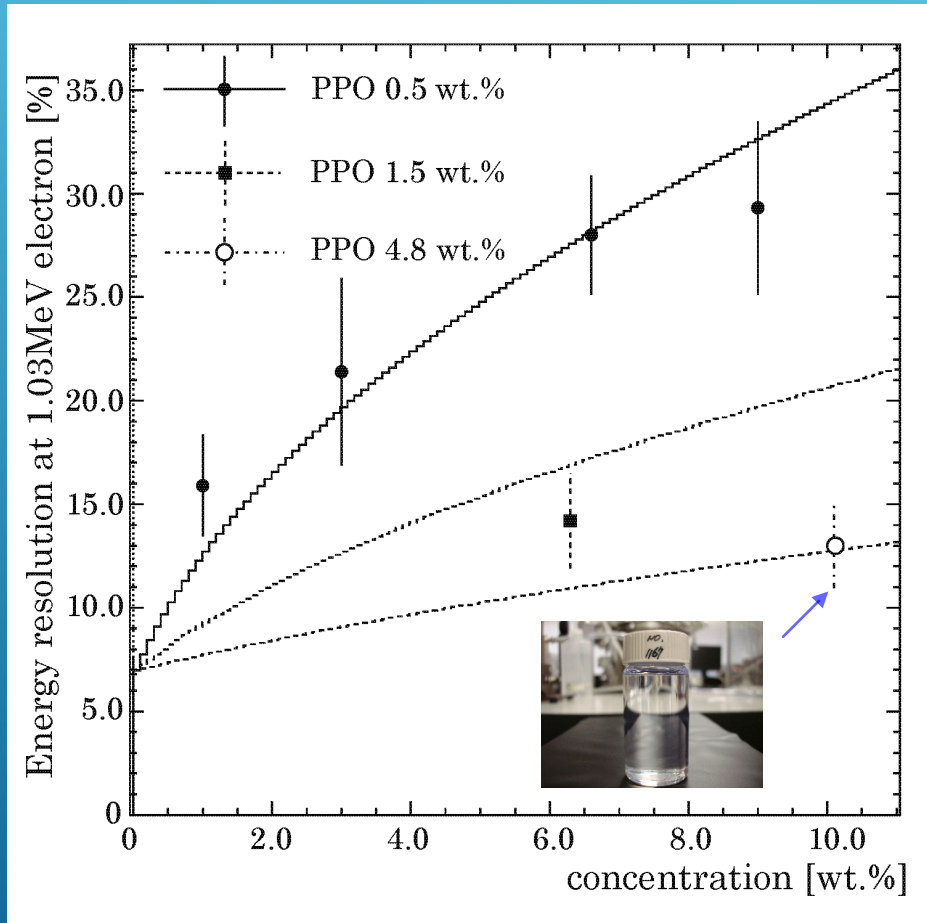
5wt.% PPO helps actually recovering the scintillation light yield.



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

Recovering the energy resolution

Measured at several conditions of PPO concentration

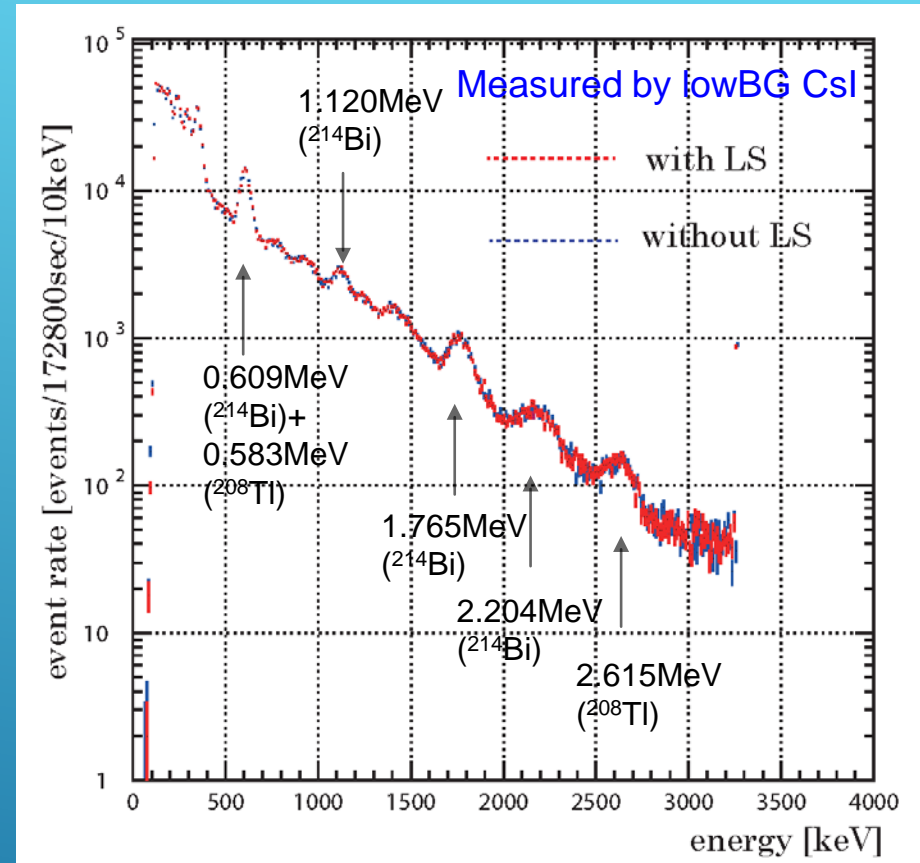


5wt.% PPO helps again the energy resolution 35% → 13%. at 10wt.% of Zr(iprac)₄.

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

Achieved goal !

Measurement of backgrounds from LS



Using subtracted # of events around 2.6MeV and 2.2MeV

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

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