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Ｘ

Supported by Grant-in-Aid for Scientific Research on Innovative Areas
No.24104501 and No.26105502

日本物理学会　2016年秋季大会

2016年9月23日

宮城教育大学教育学部　福田善之、亀井雄斗、那仁格日楽*、小畑　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

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

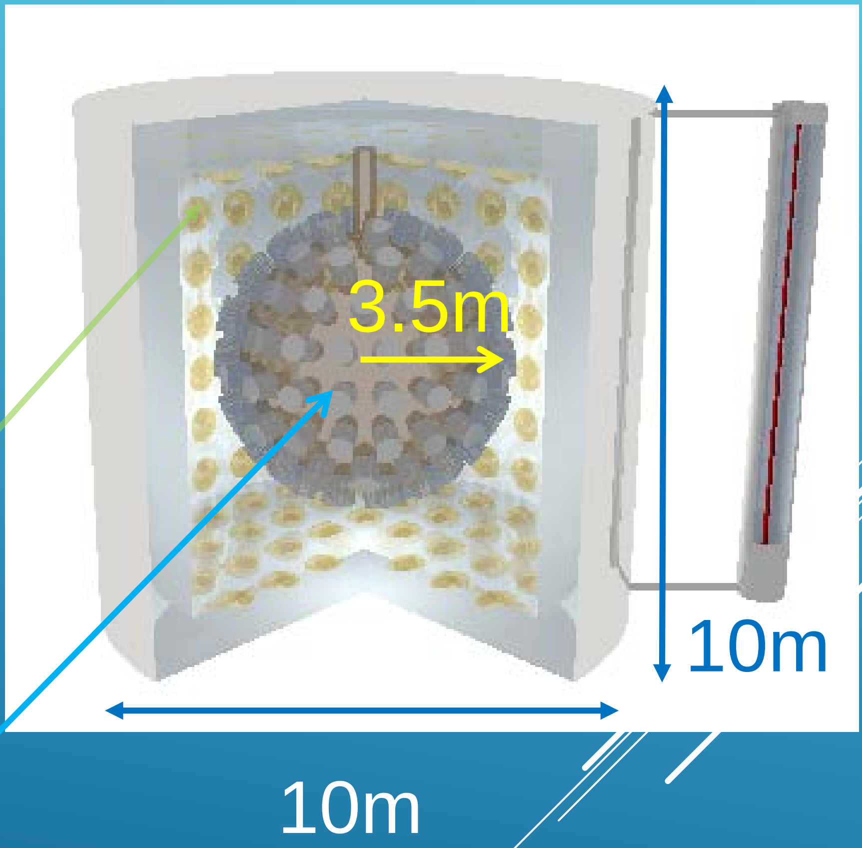
Liquid Scintillator：

(1) 10 wt.% $Zr(iprac)_4$ loaded in anisole

(2) 3.5% at 3.35MeV of energy resolution with 40% photo coverage and long attenuation length (> 6m) was obtained.

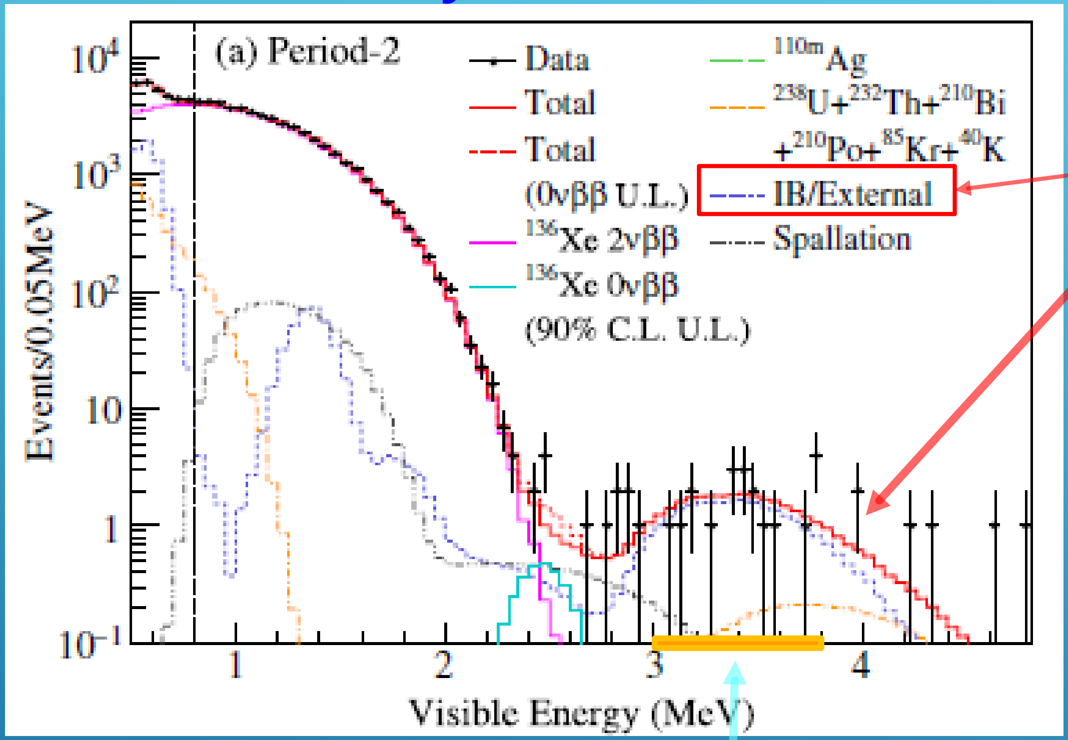
Pure water surrounding inner detector in order to veto muons and external backgrounds.

Inner detector with 40%~60% photo coverage 20" HPD PMT including 1.7ton Zirconium loaded 113 tons LS in fiducial volume. (Total vol. : 180 tons)



Backgrounds around Q-value

Measured by KamLAND-Zen



Phys.Rev.Lett. 117 (2016) 082503

^{208}Tl
In balloon

Lowering BG (^{208}Tl) < 1/20 ×KL-Zen (~1.0events/ton/year)

0νββ signal region for ^{96}Zr

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

BG reduction should be necessary.

Main backgrounds

	Period-1 (270.7 days)		Period-2 (263.8 days)	
Observed events	22		11	
Background	Estimated	Best-fit	Estimated	Best-fit
^{136}Xe $2\nu\beta\beta$	···	5.48	···	5.29
Residual radioactivity in Xe-LS				
^{214}Bi (^{238}U series)	0.23 ± 0.04	0.25	0.028 ± 0.005	0.03
^{208}Tl (^{232}Th series)	···	0.001	···	0.001
^{110m}Ag	···	8.5	···	0.0
External (Radioactivity in IB)				
^{214}Bi (^{238}U series)	···	2.56	···	2.45
^{208}Tl (^{232}Th series)	···	0.02	···	0.03
^{110m}Ag	···	0.003	···	0.002
Spallation products				
^{10}C	2.7 ± 0.7	3.3	2.6 ± 0.7	2.8
^{6}He	0.07 ± 0.18	0.08	0.07 ± 0.18	0.08
^{12}B	0.15 ± 0.04	0.16	0.14 ± 0.04	0.15
^{137}Xe	0.5 ± 0.2	0.5	0.5 ± 0.2	0.4

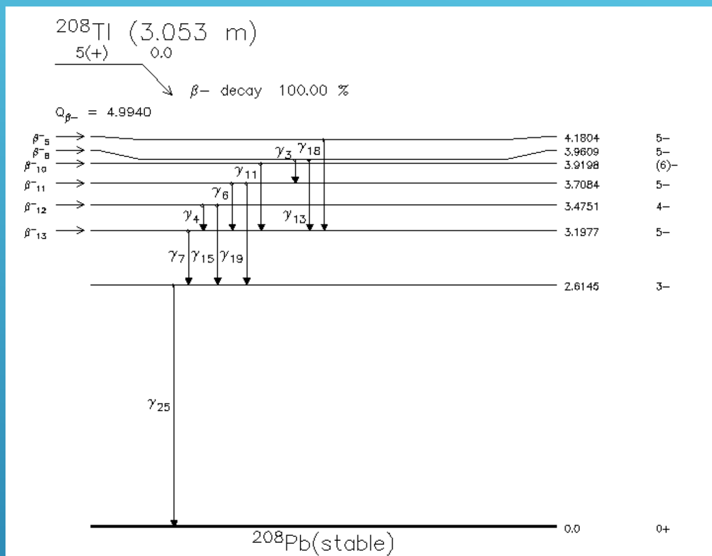
Phys.Rev.Lett. 117 (2016) 082503

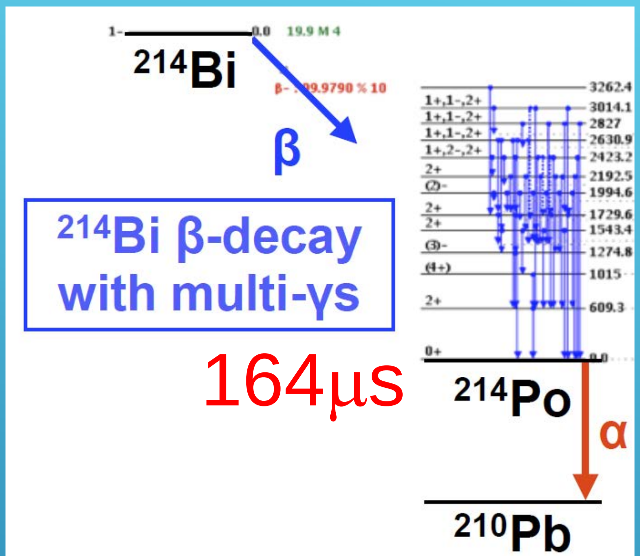
Need additional technique other than the energy spectral shape obtained by scintillation lights in order to reduce remaining backgrounds.

Decay mode

^{208}Tl : β and 2.6MeV γs

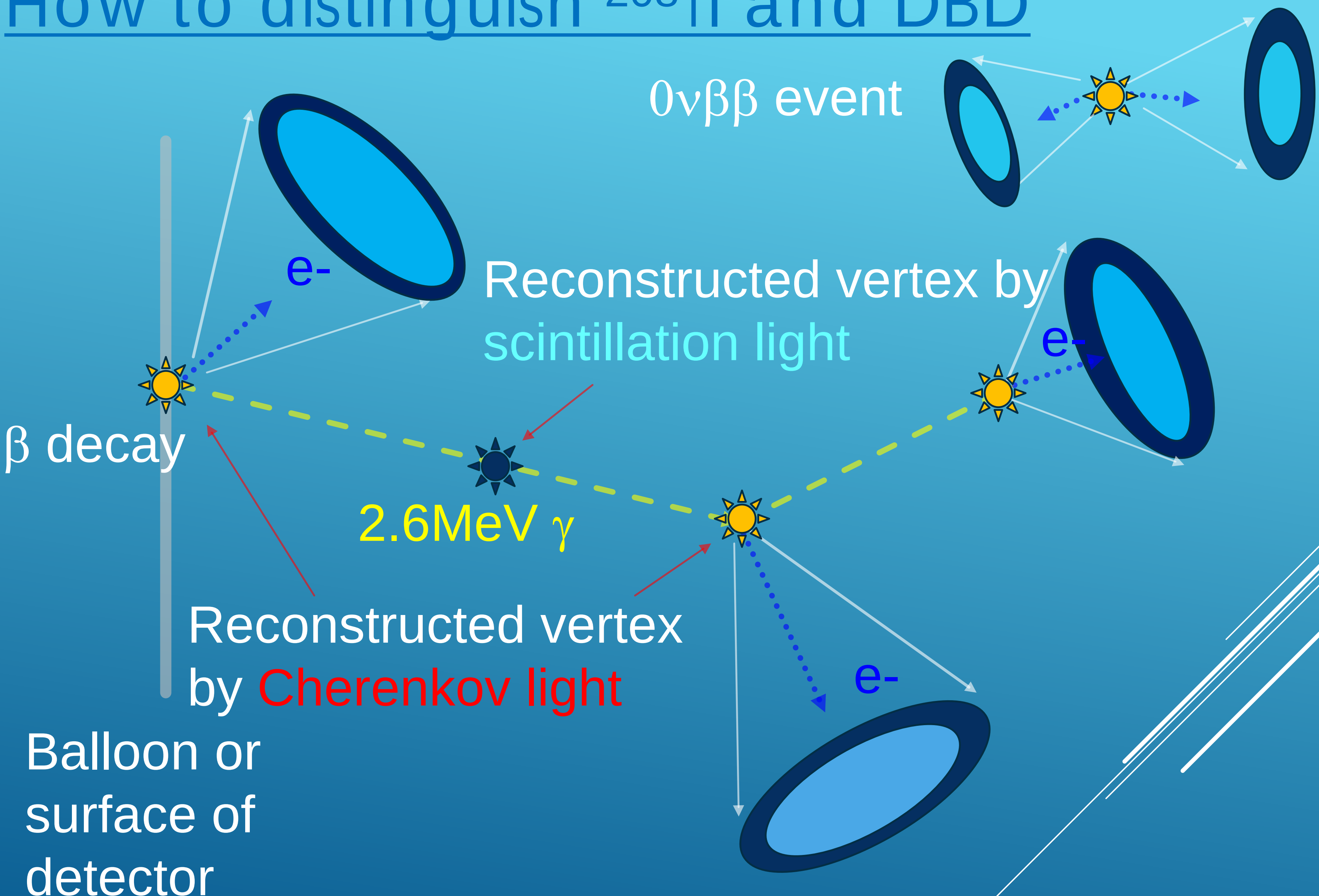
^{214}Bi :β and multi γs (609keV,1.12MeV)



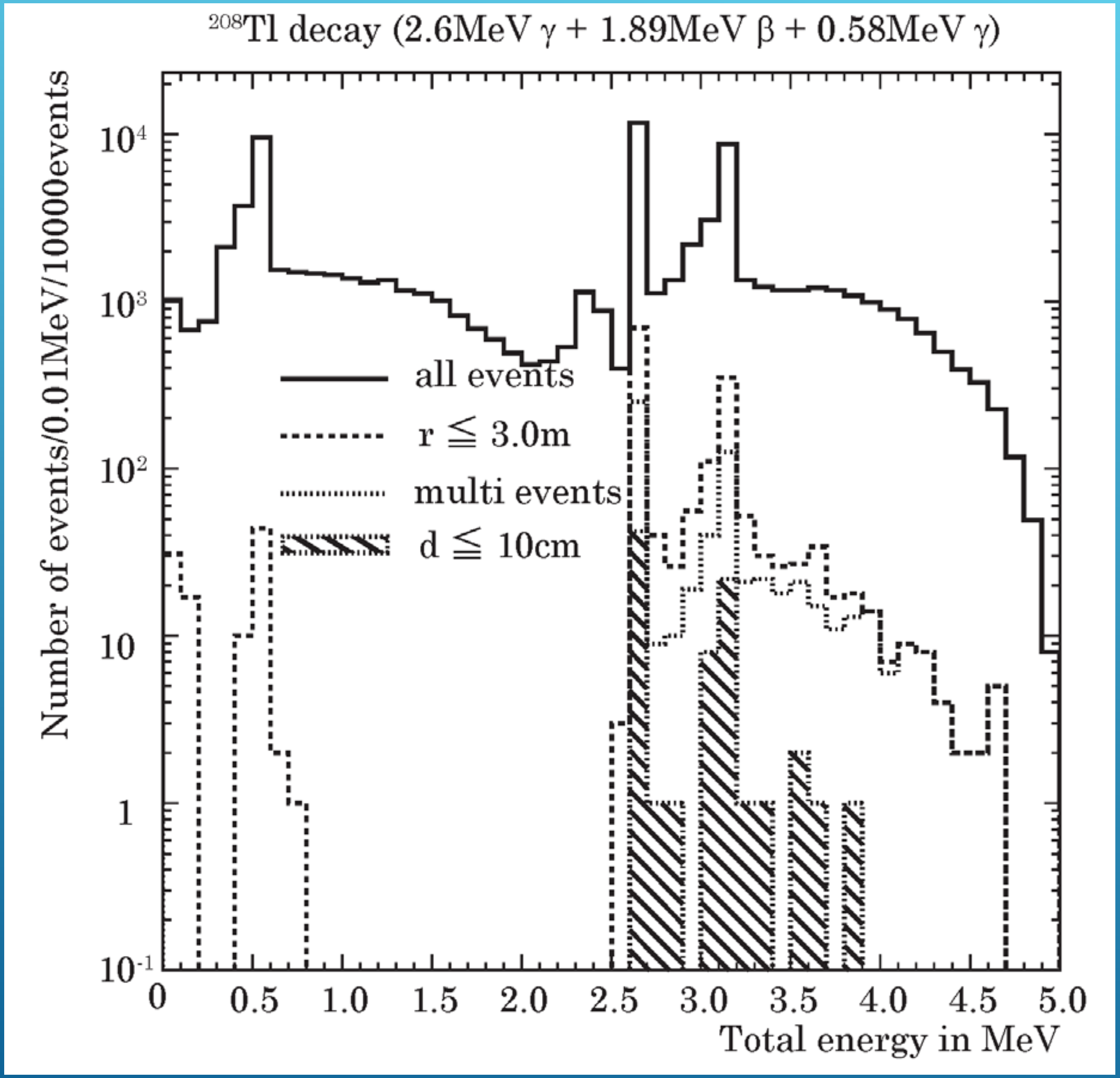


- Multiple events
  - β and gammas (2.6MeV + etc) for ^{208}Tl
  - β and complicated gammas, and α for ^{214}Bi
- Mis-reconstruction and mis-identification
  - Reconstructed as one event even if multiple events
  - Caused contamination in fiducial volume

How to distinguish ^{208}Tl and DBD



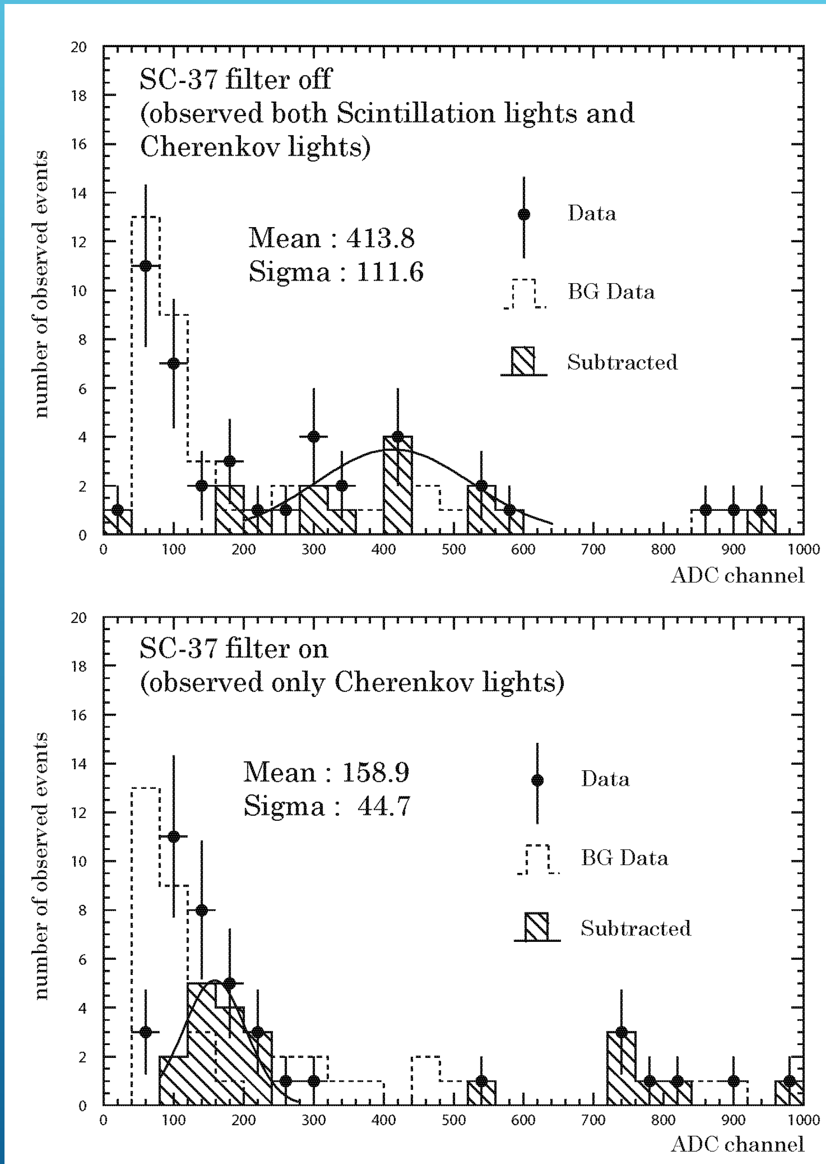
Simulation of ^{208}Tl decay

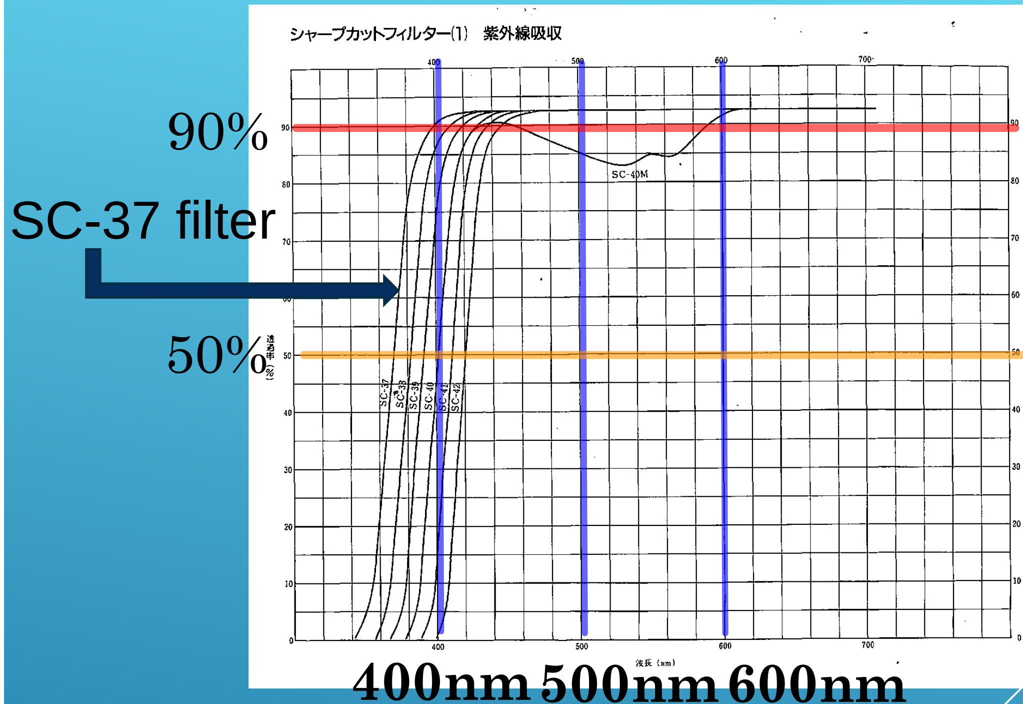


1) E : 3.0-3.7MeV
17925 events
2) Fiducial volume
628 events
3) Multi events
263 events
3) Closer events
(d≦10cm)
35 events

1/20 BG reduction could be achieved.

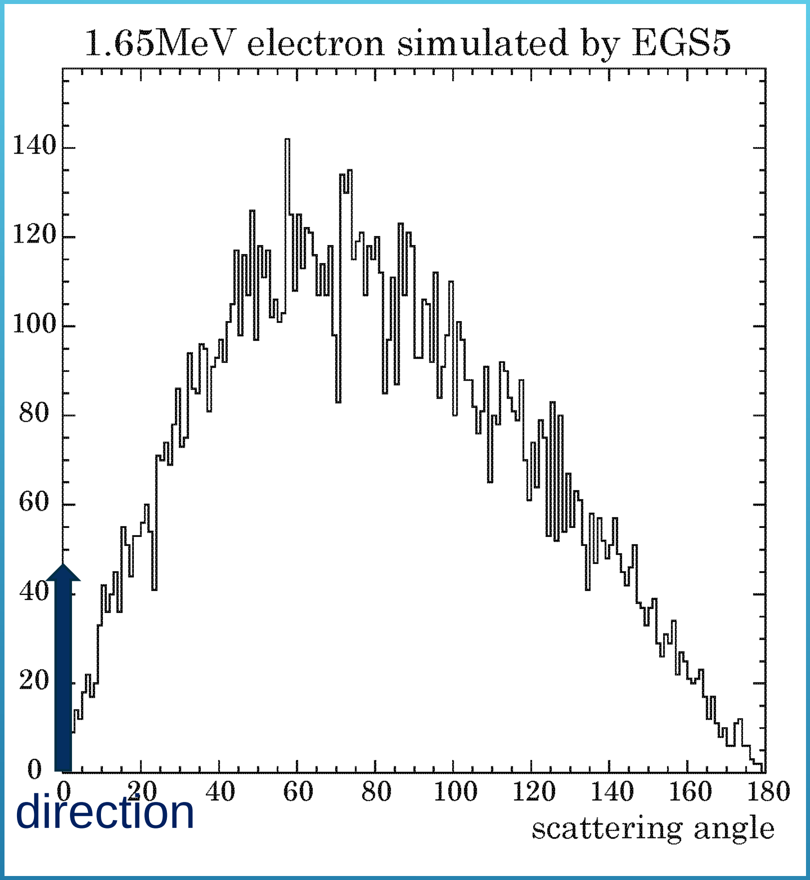
Light yield of Cherenkov lights

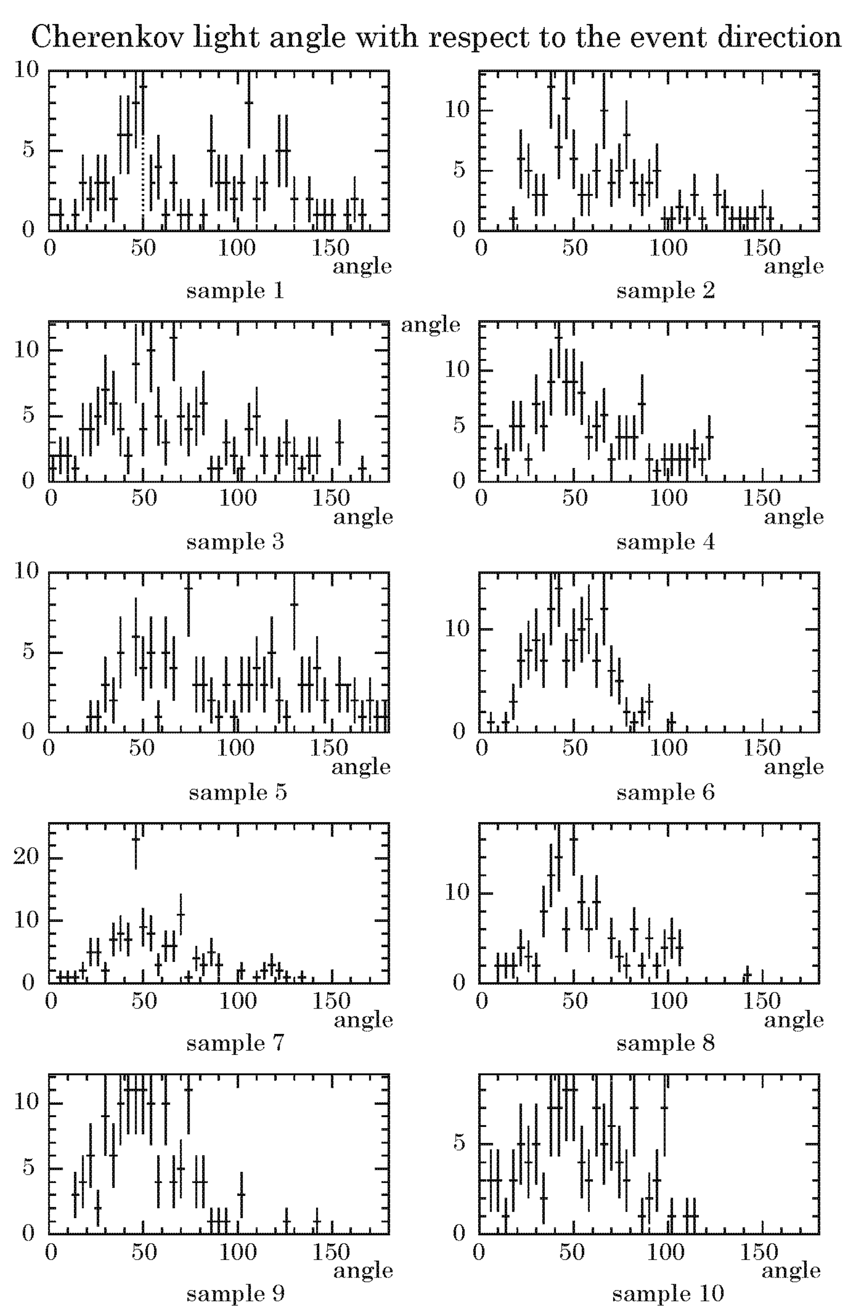




$$\frac{\text{Cherenkov light yield } (\lambda > 400\text{nm})}{\text{Scintillation light yield of anisole}} = \sim 0.02 \equiv \sim 200 \text{ photon/MeV}$$

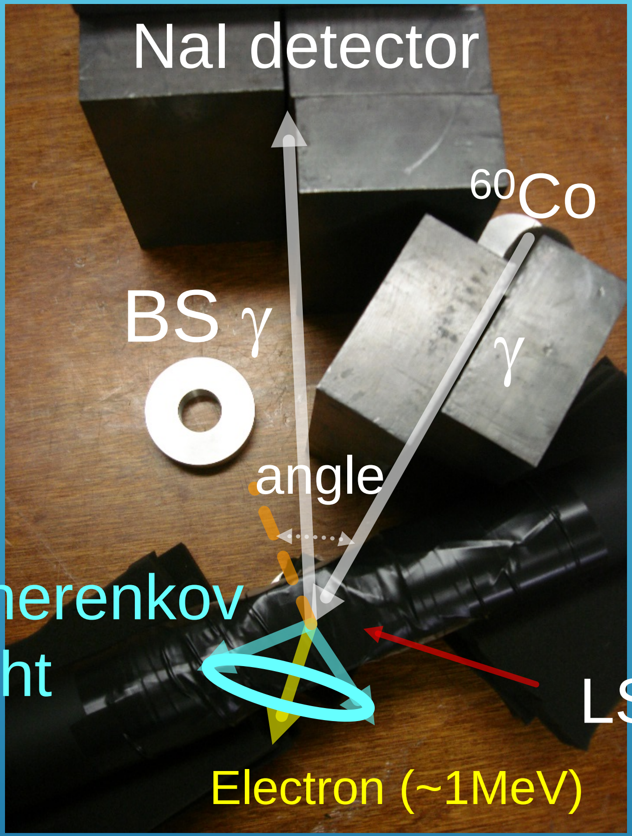
Multiple scattering

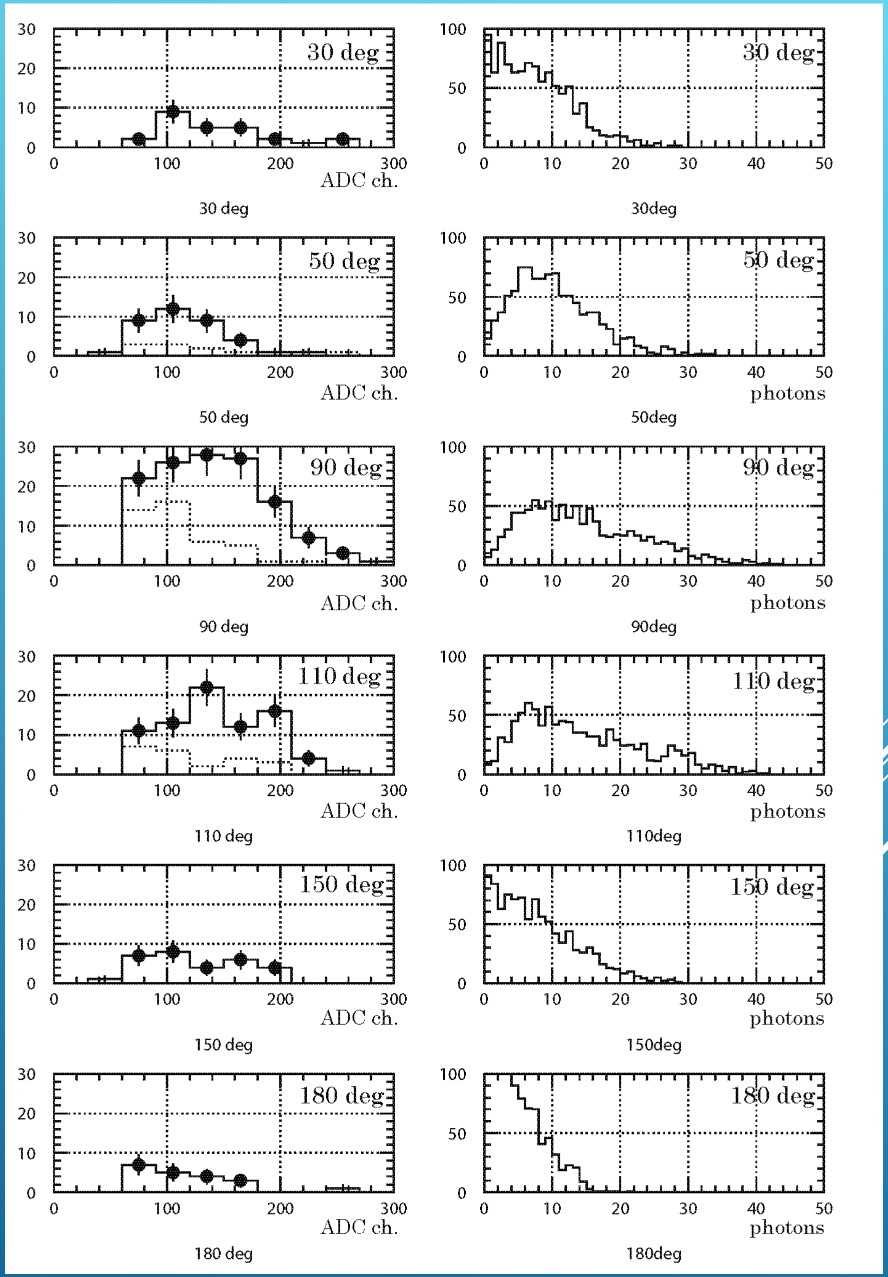




Even though multiple scattering of electrons, Cherenkov light looks have some clusters in the directional distribution.

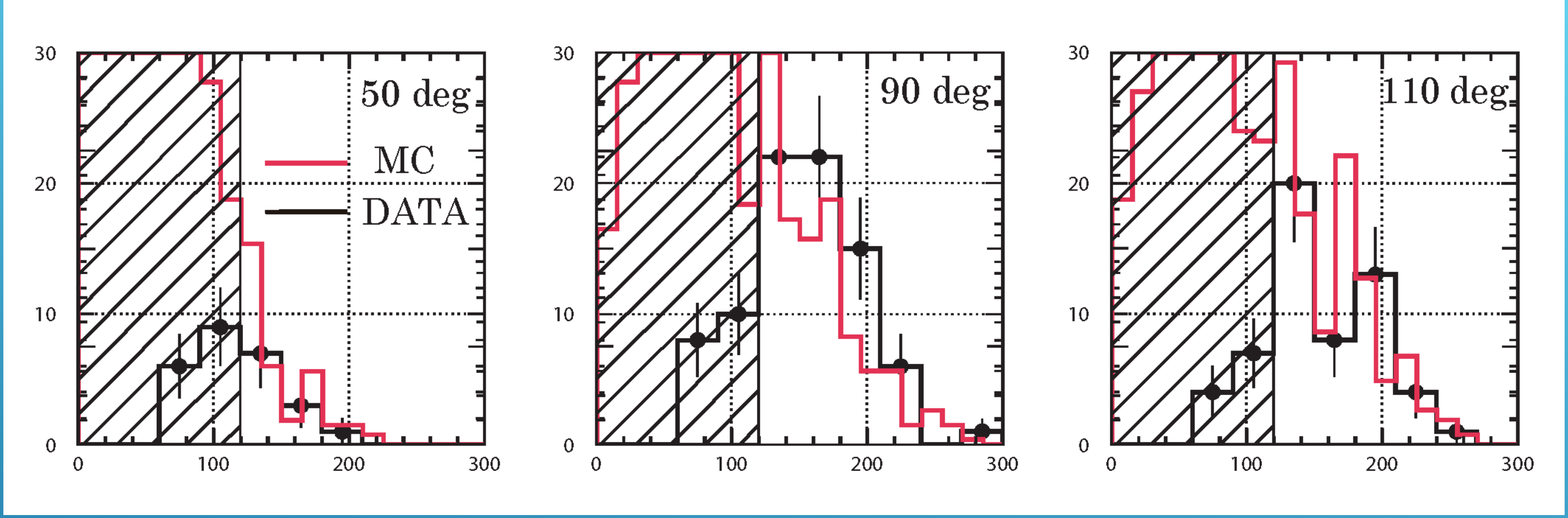
Angular dependence





Data indicated same angular dependence as MC simulation of Cherenkov lights with EGS5.

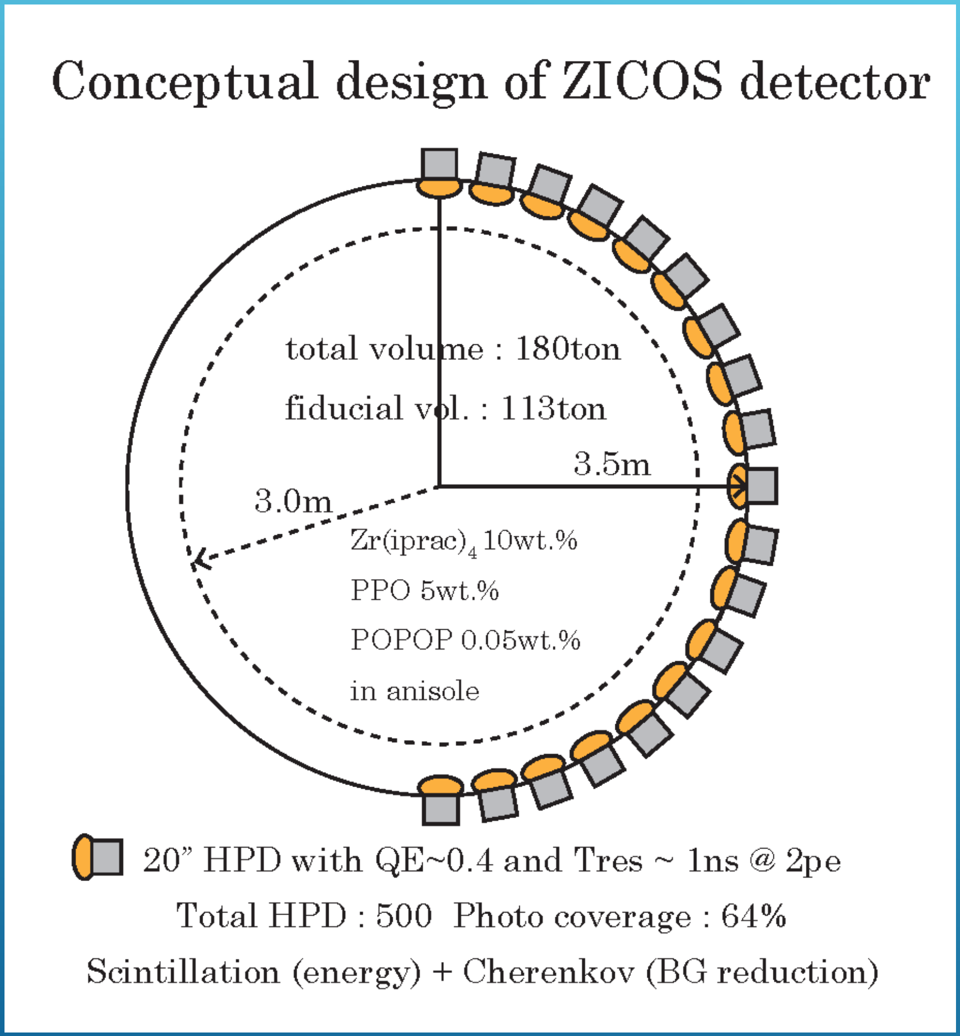
Comparison between MC and Data



EGS5 based MC almost reproduced the angular dependence of data.

Cherenkov lights emitted by ~1MeV electron really have a directionality.

Design of ZICOS detector



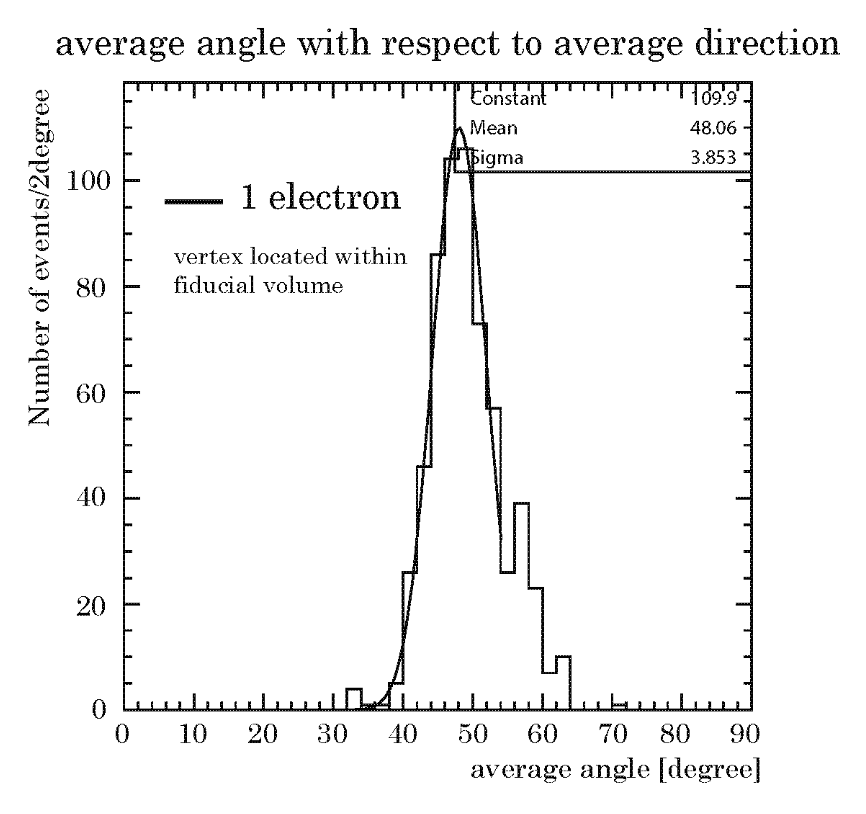
Detector :

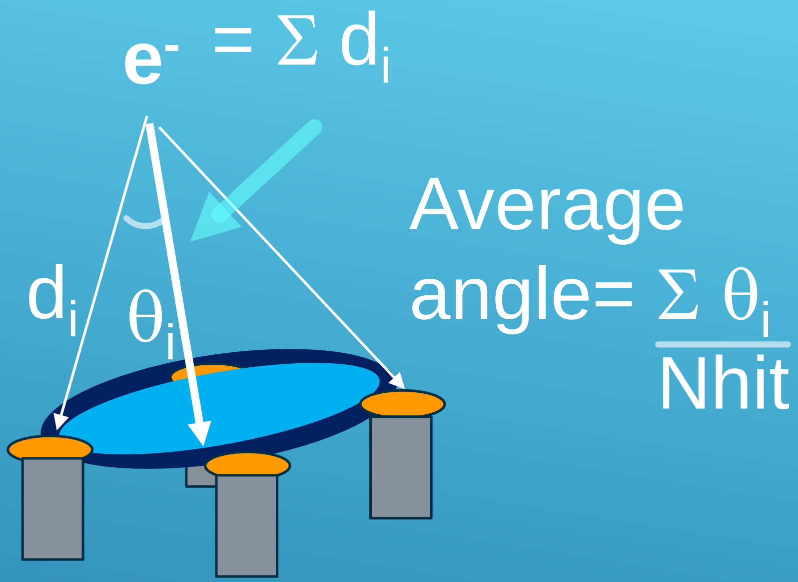
1) 180tons LS : 10 wt.% $Zr(iprac)_4$ with PPO/POPOP in anisole Need 500 of 20" HPD with high QE ~0.4 and Tres (~1ns @ 2pe) = 64% coverage

Expected performance :

1) Energy resolution ~2.8%@3.35MeV
2) $T_{1/2}(0\nu\beta\beta) > 10^{26}$ years, if both 1/20 BG reduction and 50% ^{96}Zr enrichment could be achieved.

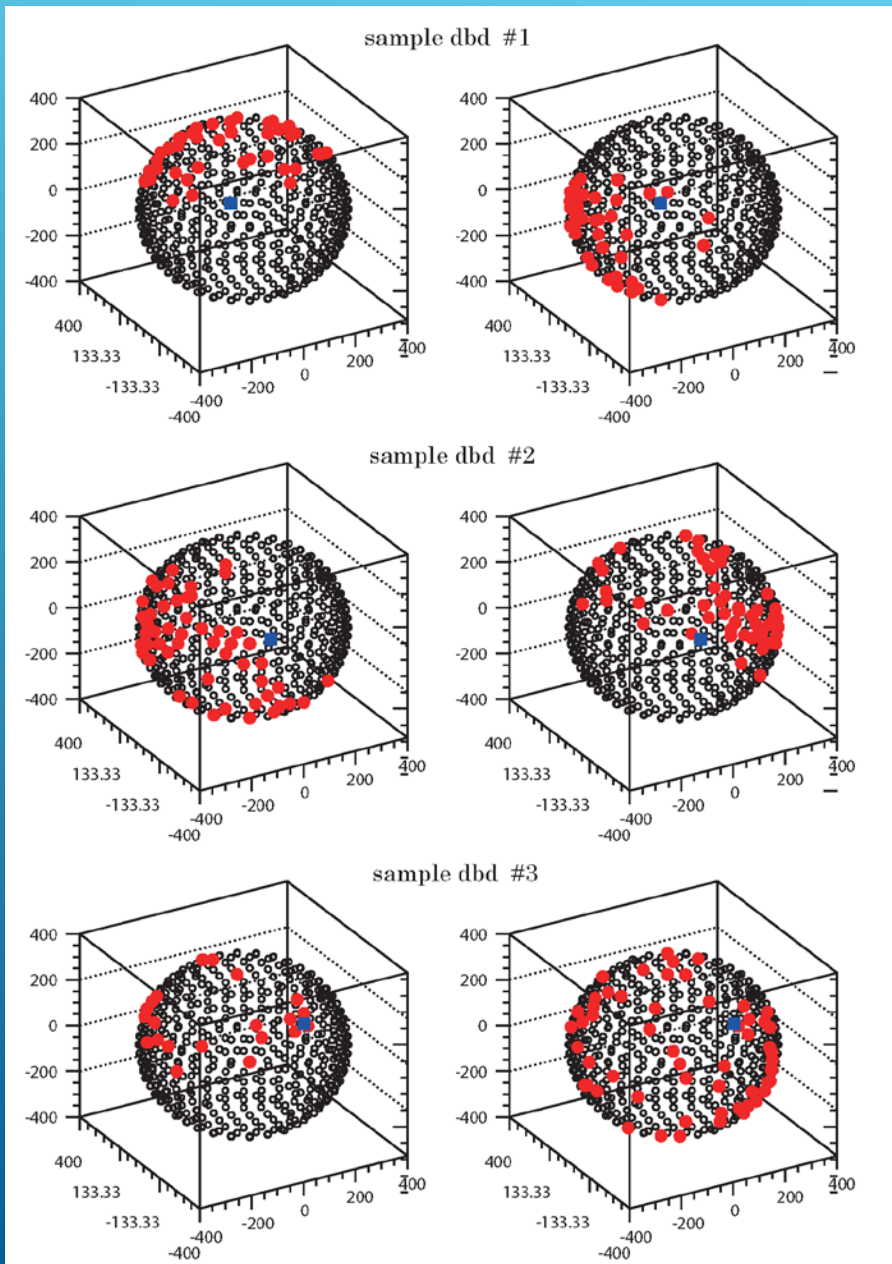
Average angle distribution

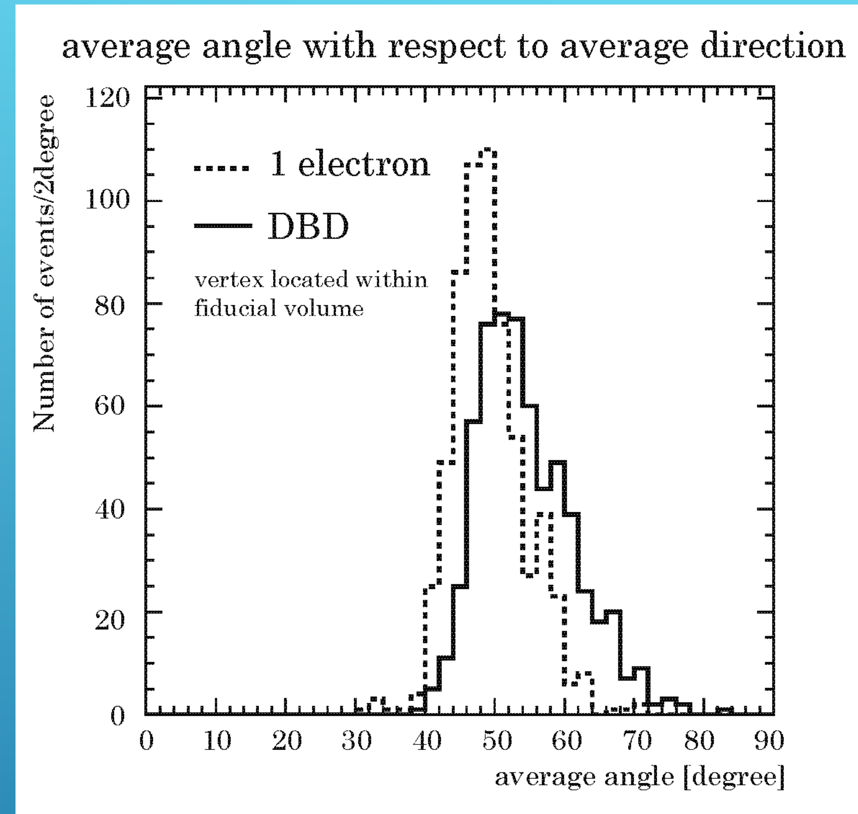




Average angle with respect to averaged direction for single electron seems to have a peak at 48 degree which is almost same as Cherenkov angle.

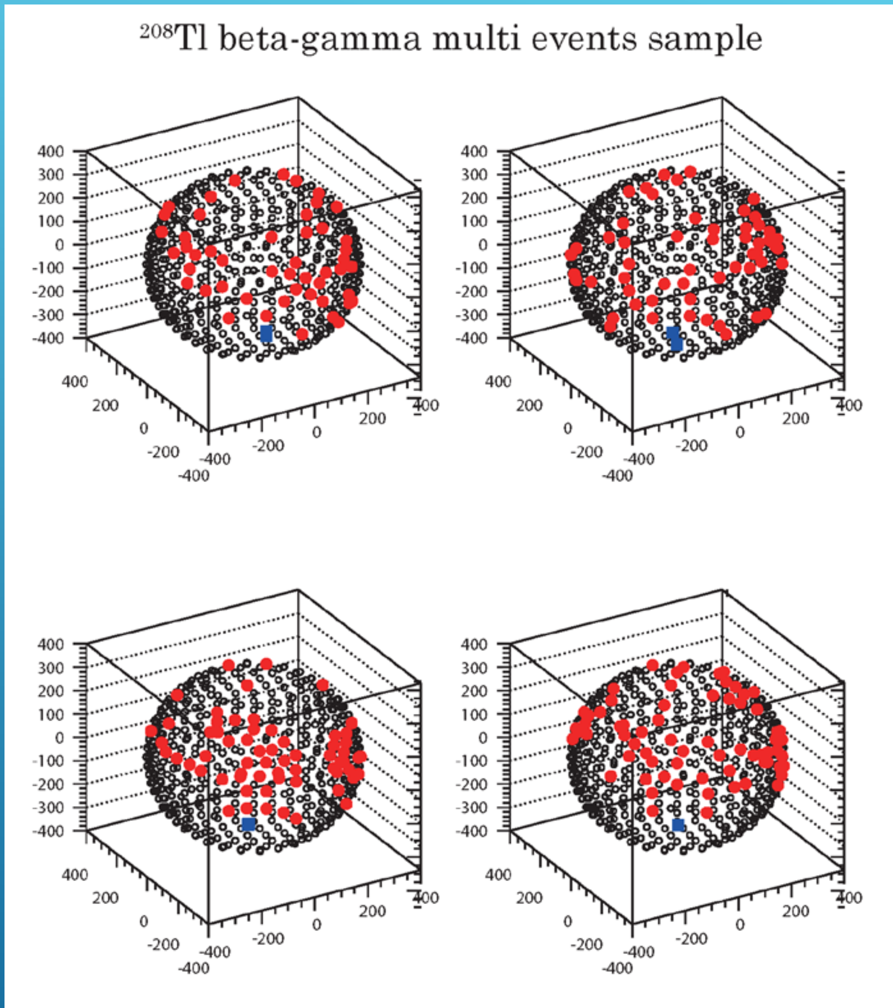
Hit pattern of DBD (opposite and half E)

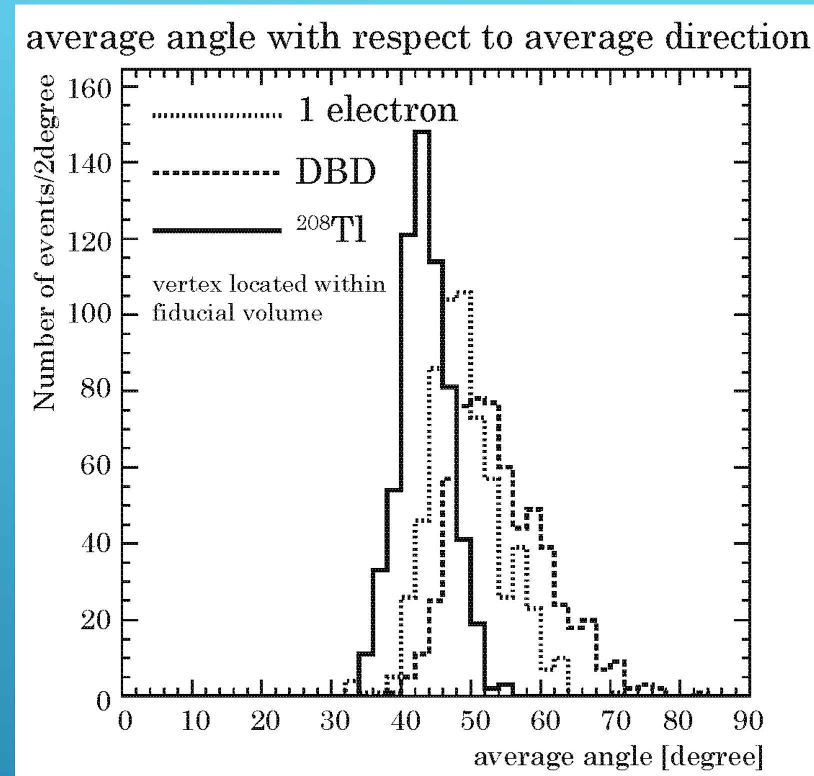




Multi events from DBD tend to have a slightly larger angle than single electron.

Hit pattern of ^{208}Tl (2.6MeVγ+ β+γ)

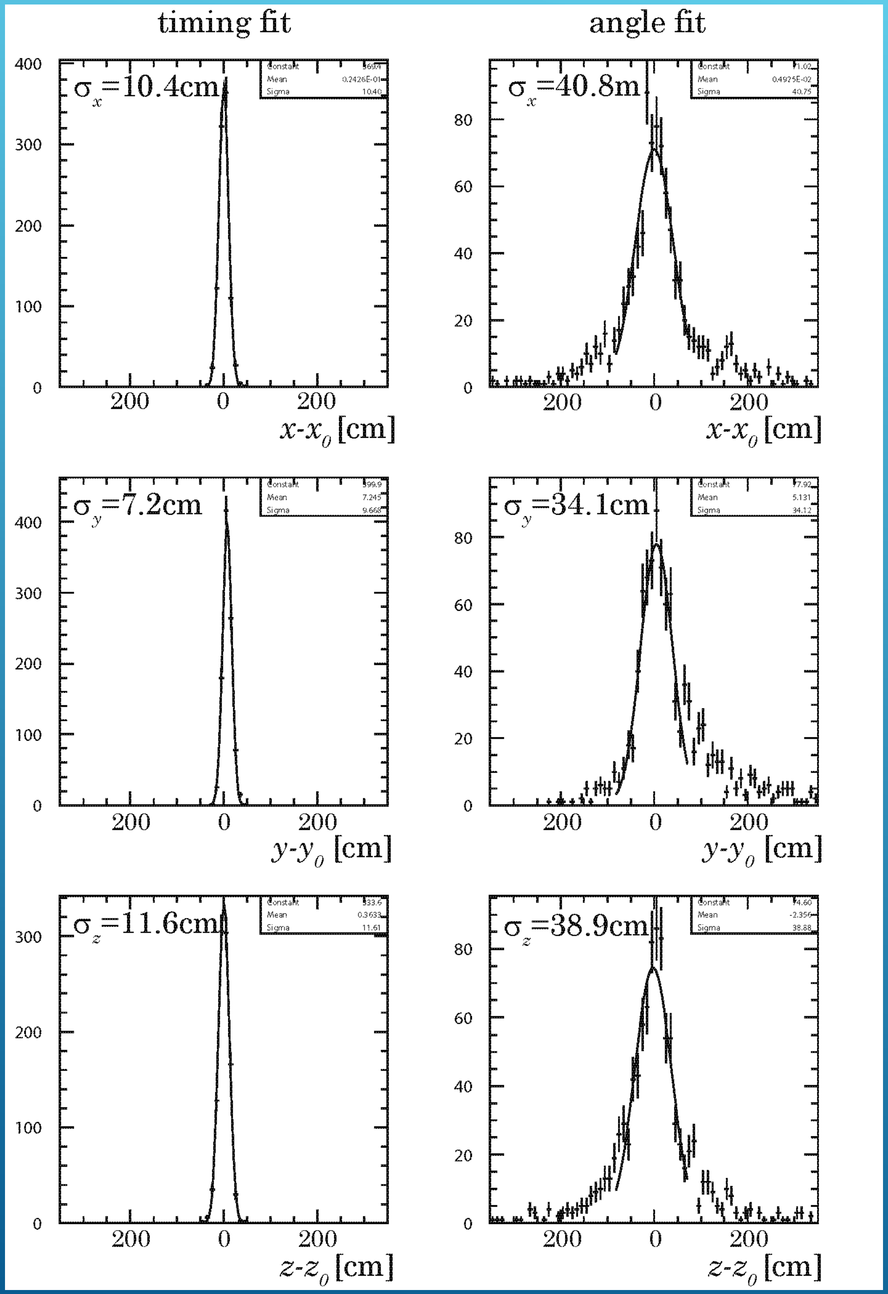




Multi events from ^{208}Tl tend to have a smaller angle than DBD or single electron, even vertex obtained by scintillation.

Hit pattern could be used for BG reduction.

Reconstructed vertex resolution



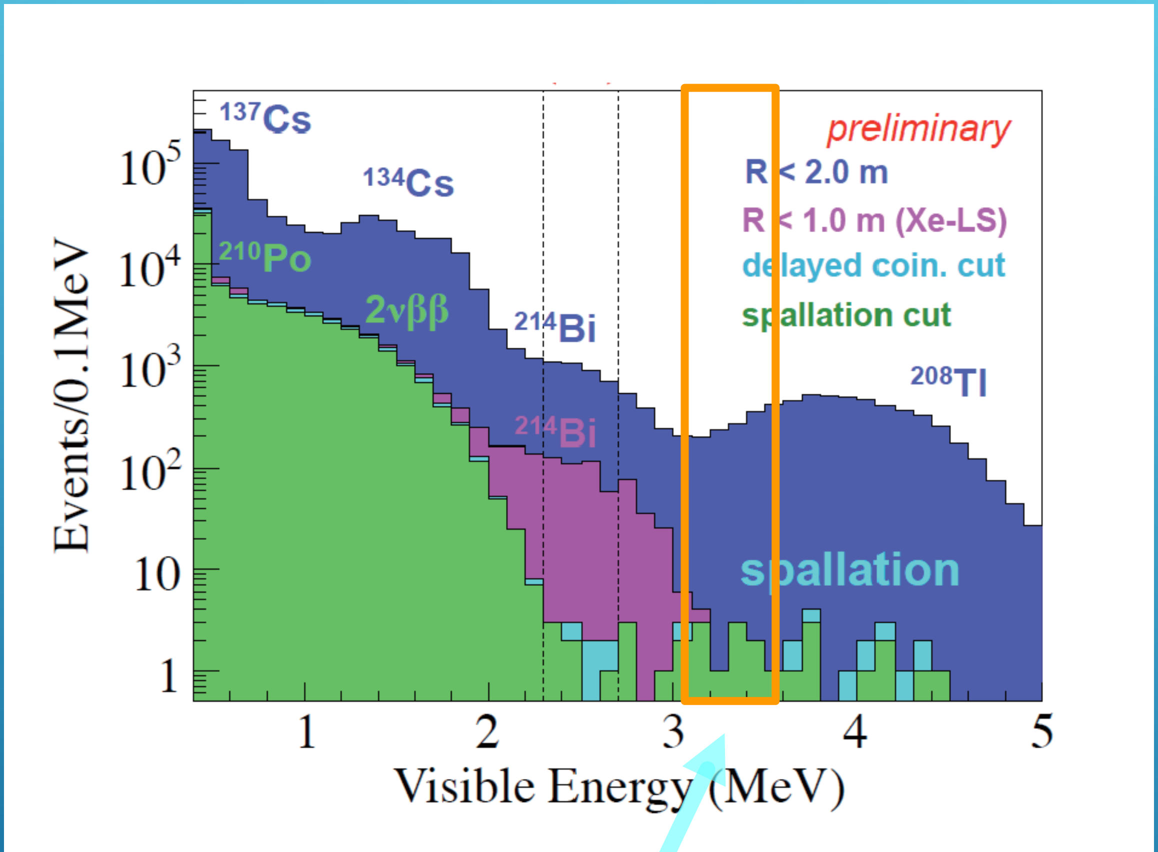
- HPD should have better QE ~0.4 and timing resolution ~1ns @ 2pe than HK HPD in order to reconstruct the vertex position with σ～10cm.
- Could reconstruct the vertex position using even only hit pattern, however need more smart method to get better resolution.

Summary

- **Further 1/20 reduction of ^{208}Tl (and ^{214}Bi) than KLZ could be realized by identifying as multi events.**
- **Simulated Cherenkov lights with EGS5 and could reproduce the angular dependence which is seen by data.**
- **Conceptual design for ZICOS detector with 20" HPD with 64% photo coverage was presented.**
- **Need to develop HPD with higher QE (~40%) and better timing resolution (~1ns @ 2pe) in order to obtain vertex resolution $\sigma \sim$10cm.**
- **Need smart methods both to recognize multi events whether DBD or BG and to get a vertex position using hit pattern.**

BACKUP

Backgrounds around 3.35MeV



I.Shimizu@Neutrino2014

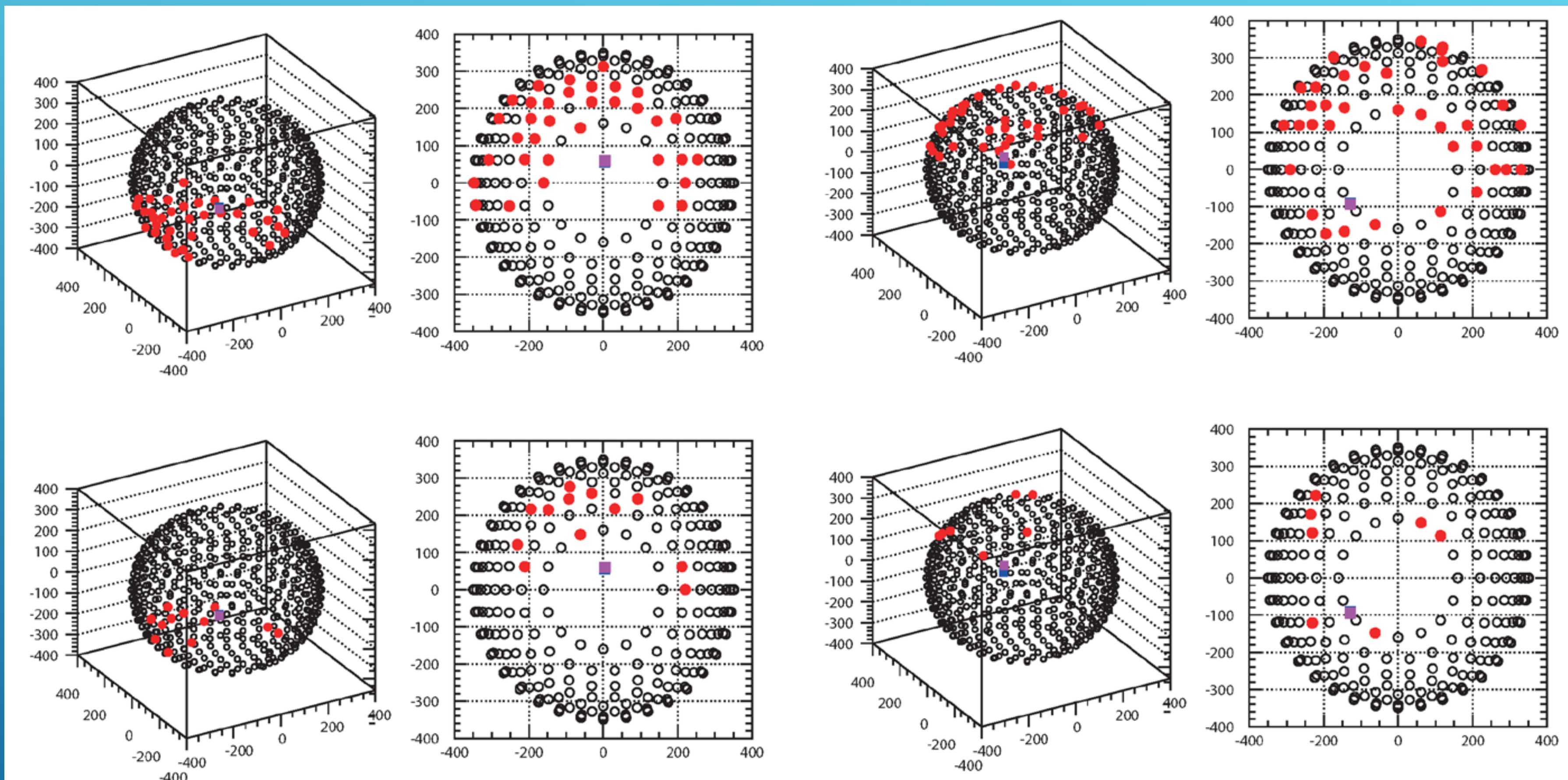
$0\nu\beta\beta$ signal region for ^{96}Zr

^{214}Bi contaminates within 1m inner volume, but most of events could be removed by Bi-Po tagging. Also ^{214}Bi is not serious BG in terms of signal region for ^{96}Zr

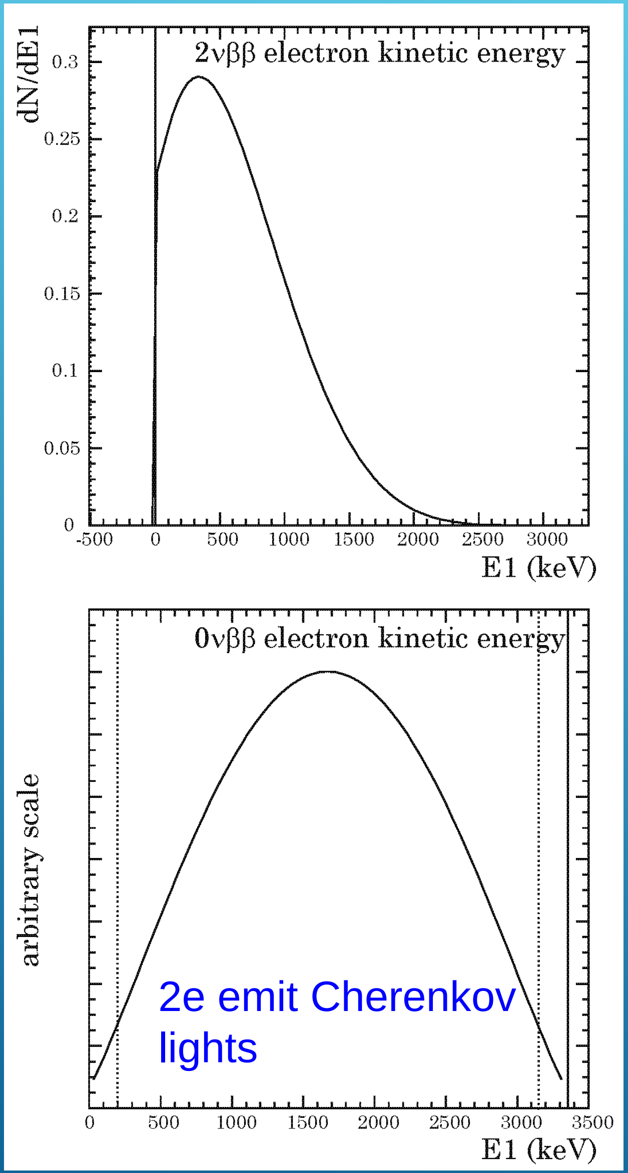
Most serious BG is ^{208}Tl for ^{96}Zr

Hit pattern of Cherenkov lights

Simulated by EGS5 (kinetic energy 1.65MeV)



Electron kinetic energy spectrum



For calculation of 2νββ,

$$\frac{d\omega}{dk_1 dk_2 d\cos\theta} \sim \mathcal{F}(Z,\varepsilon_1)\mathcal{F}(Z,\varepsilon_2)k_1^2k_2^2(W_0-\varepsilon_1-\varepsilon_2)^5(1-\beta_1\beta_2\cos\theta)$$

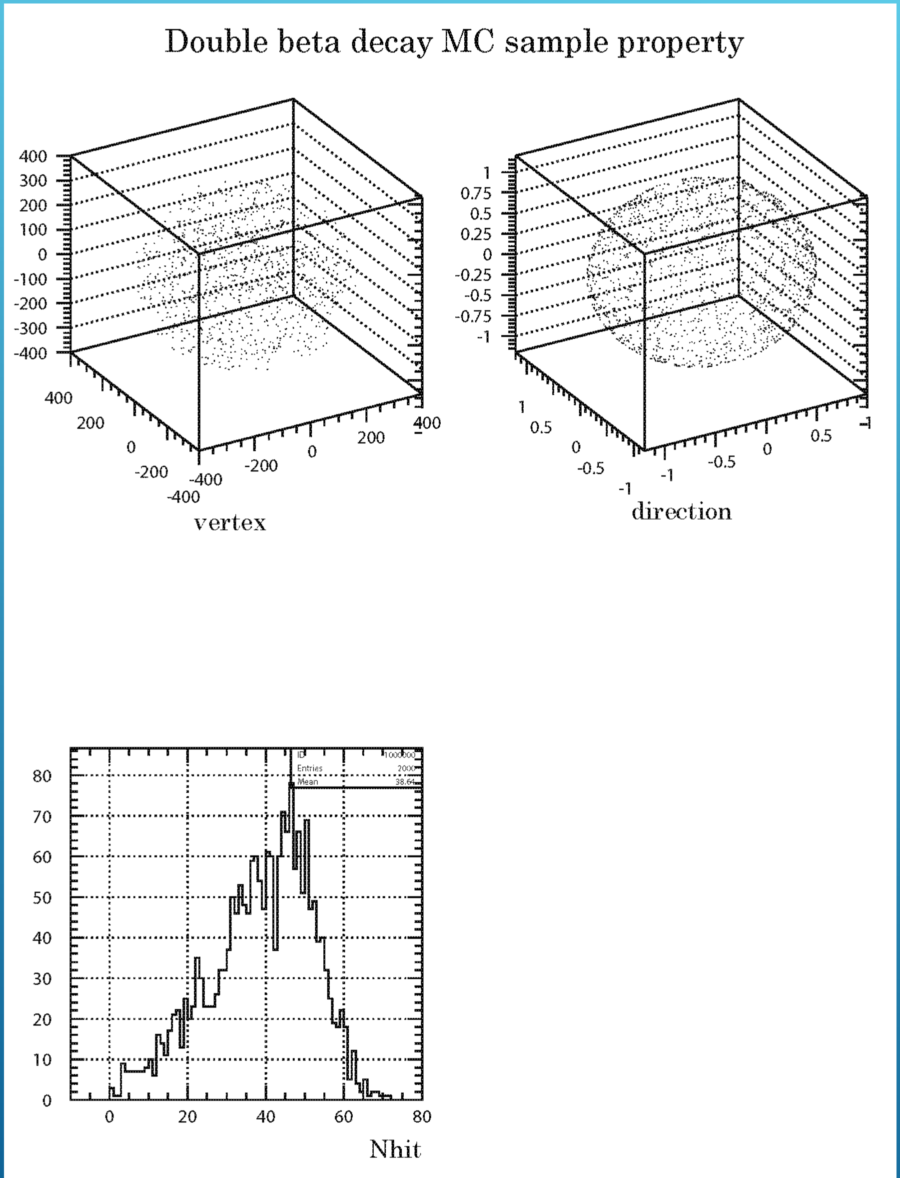
k_i, electron momenta
$\varepsilon_i = \mathrm{sqrt}(k_i^2+m_e^2)$: electron energy
$W_0=Q+2m_e$: total release energy
Q : Q value m_e: electron mass
θ : opening angle $\mathcal{F}$: Fermi func.

ε_i can generate independently within energy conservation.

For calculation of 0νββ,

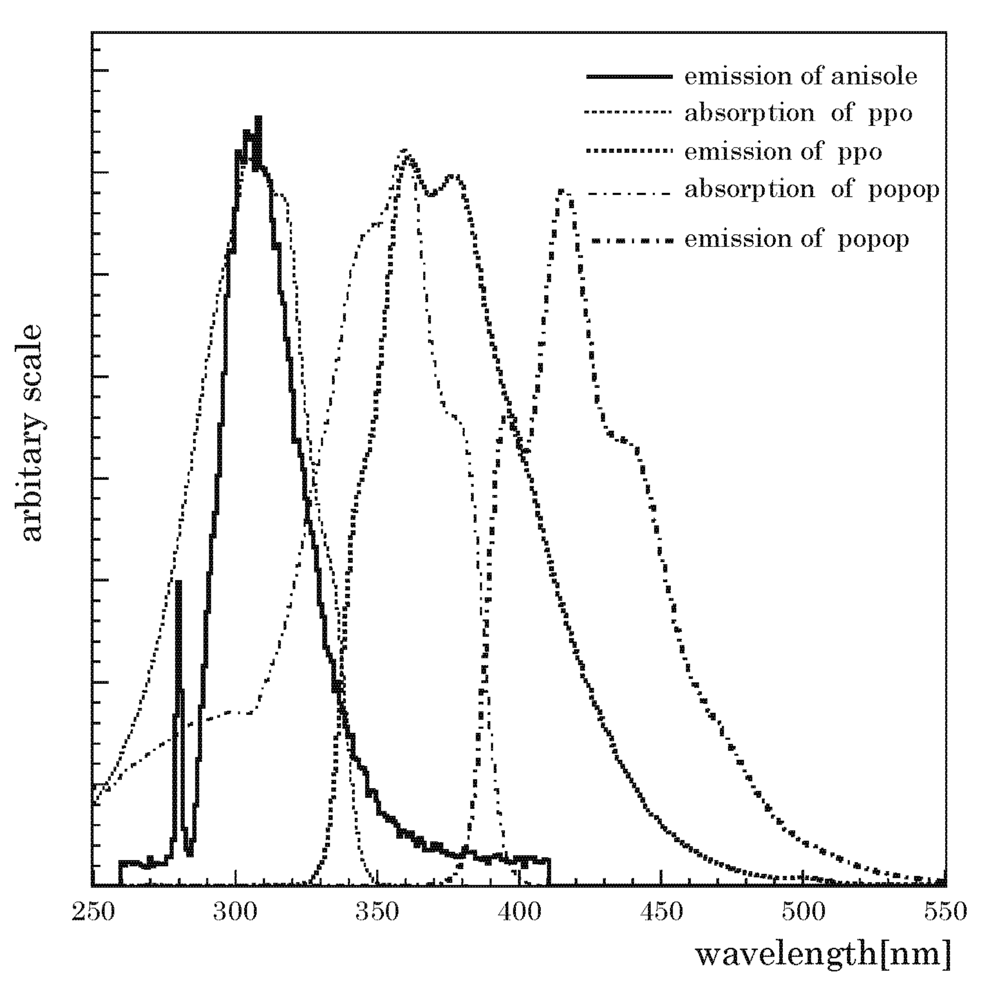
Same calculation but ε_i can only generate with $\varepsilon_1+\varepsilon_2=W_0$.

Basic distribution of DBD MC sample



- Two electrons were generated at same vertex position with opposite direction.
- Each energy is 1.65MeV.
- Cherenkov photons were generated at each EGS ausgab subroutine with 200 photons/MeV.
- QE and time resolution were assumed by 0.4 and 1ns@2pe for HPD.

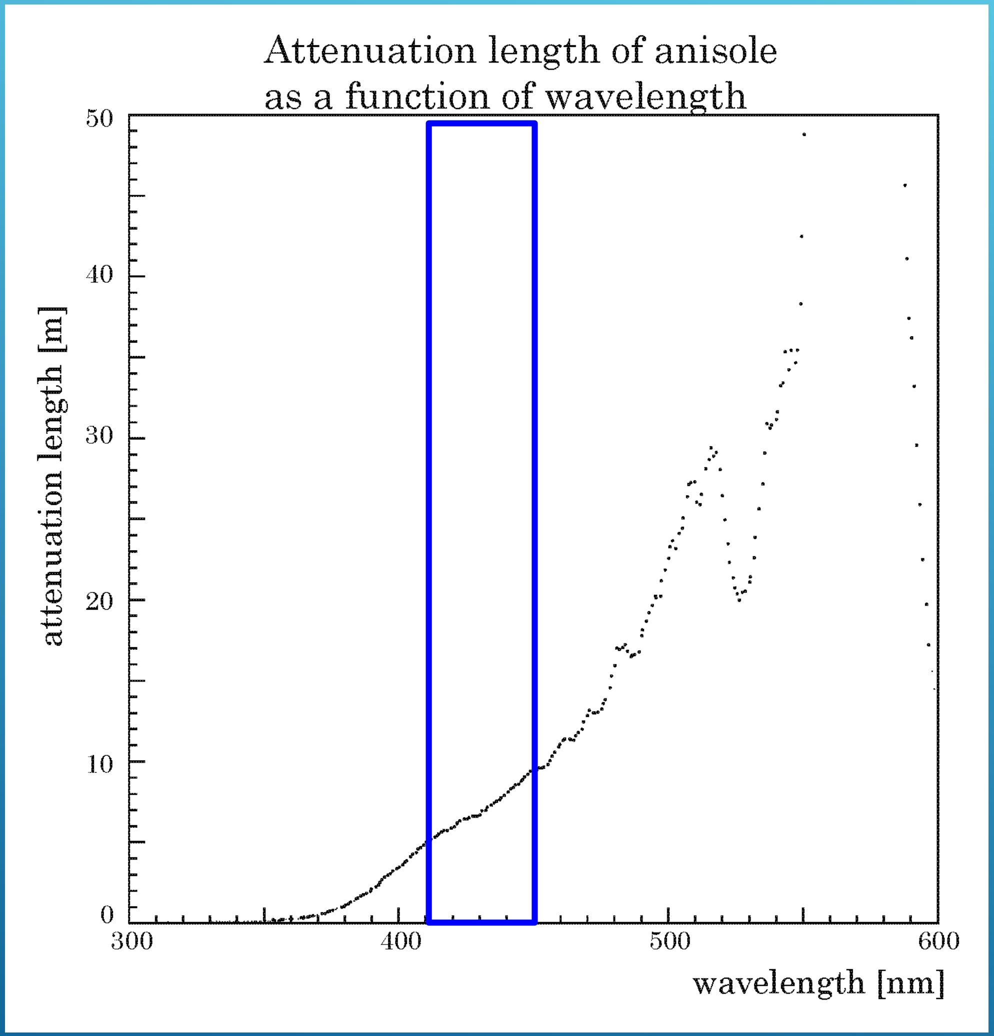
Emission and absorption spectra for solvent and solute in standard cocktail



PPO absorbed most of emission lights from anisole.

Effectively the energy was transferred to the secondary scintillator.

ATTENUATION LENGTH OF ANISOLE

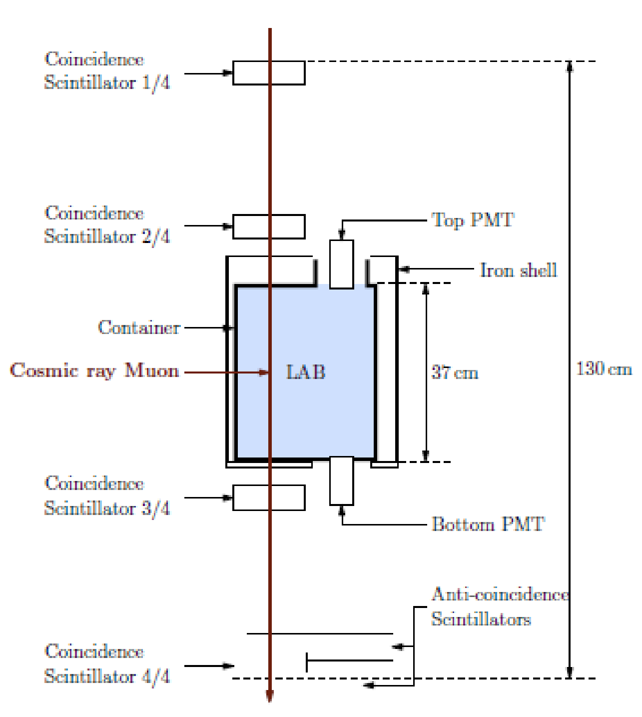


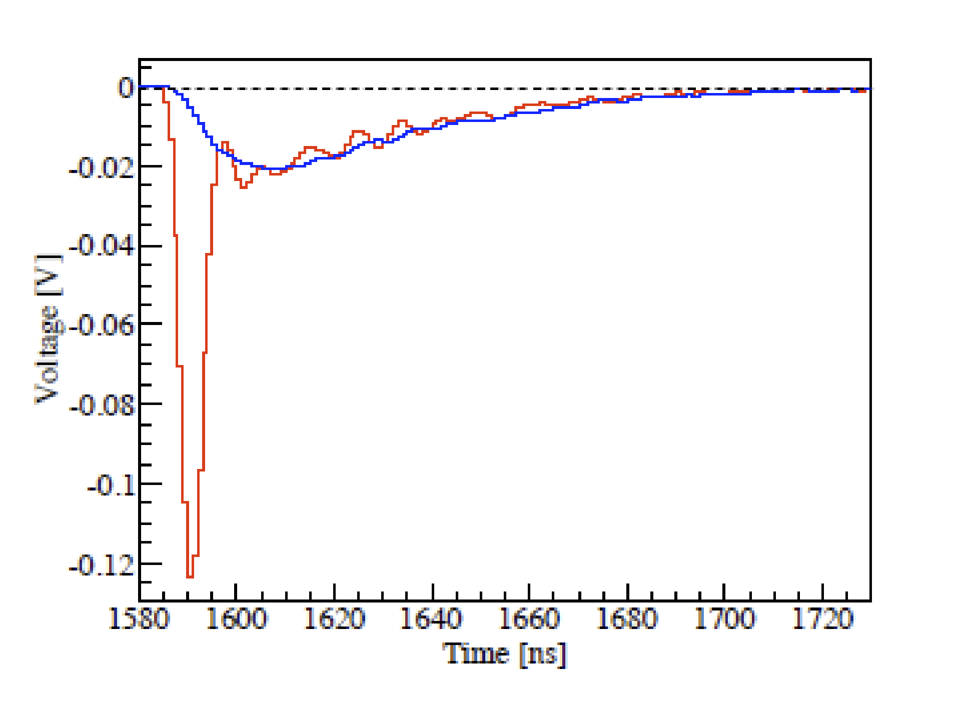
Attenuation length of scintillation light from POPOP (~450nm) was obtained as ~6m.

No problem for radius of ZICOS detector.

How to separate Cherenkov and scintillation

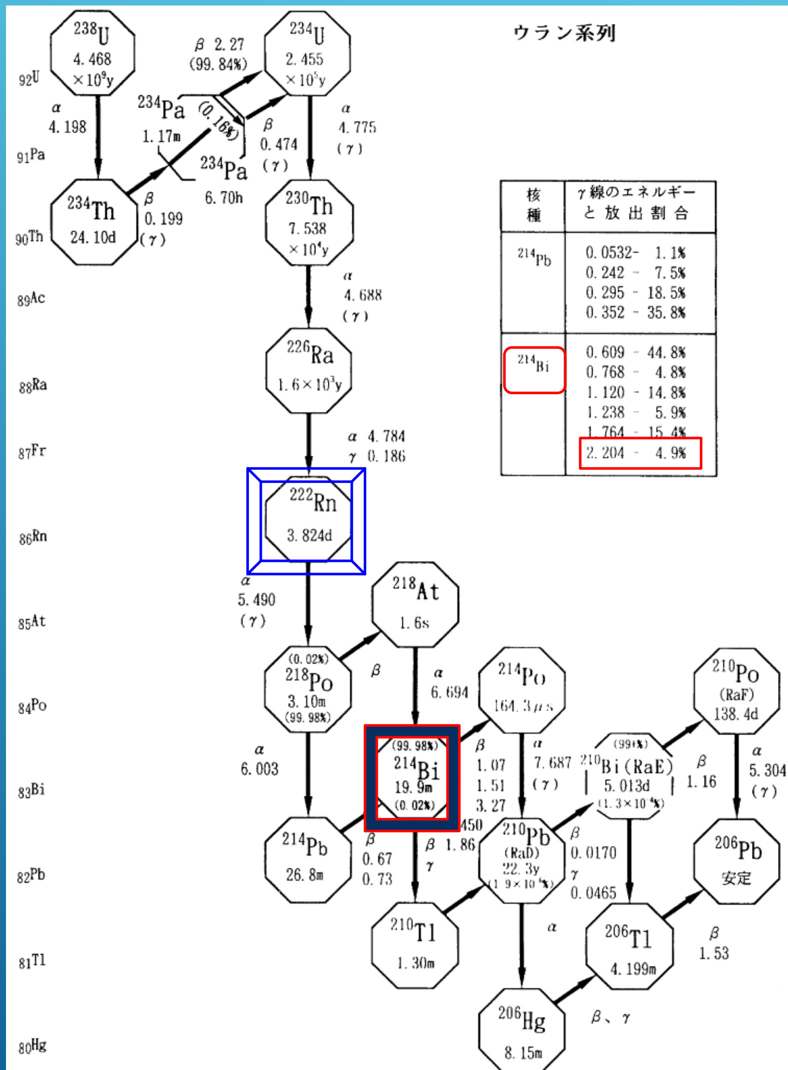
Separation of scintillation and Cherenkov lights in PMT signal using waveform of FADC. C.Shaomin et al. arXiv:1511.09339

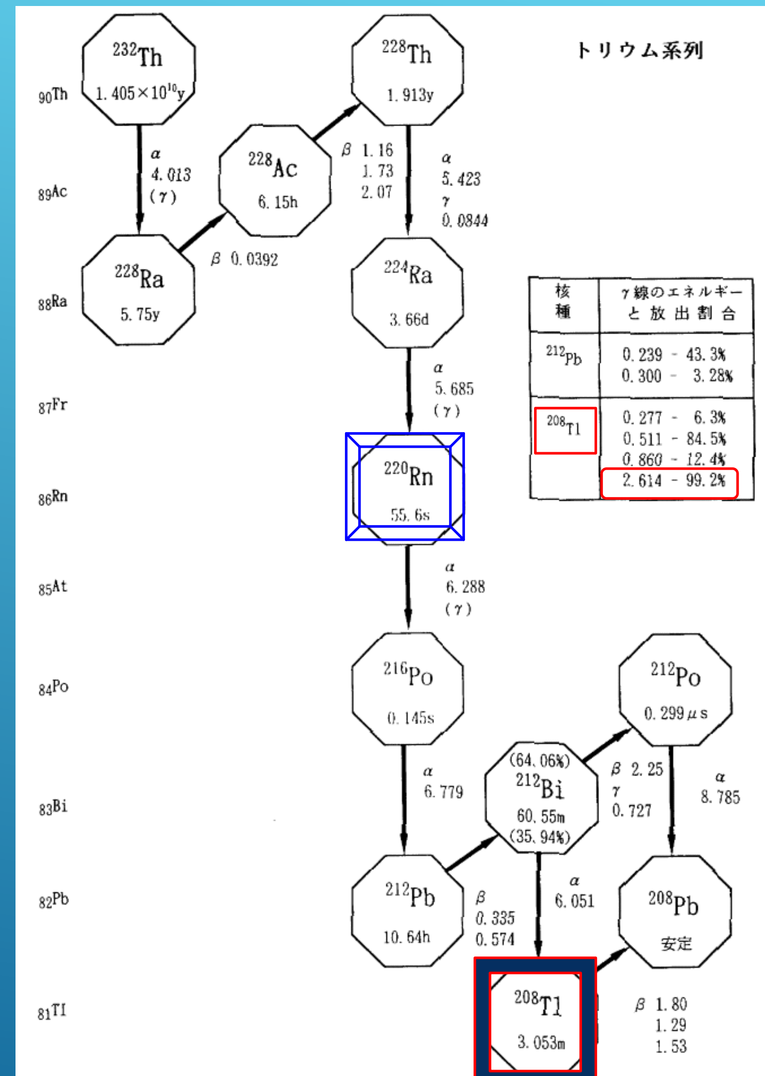




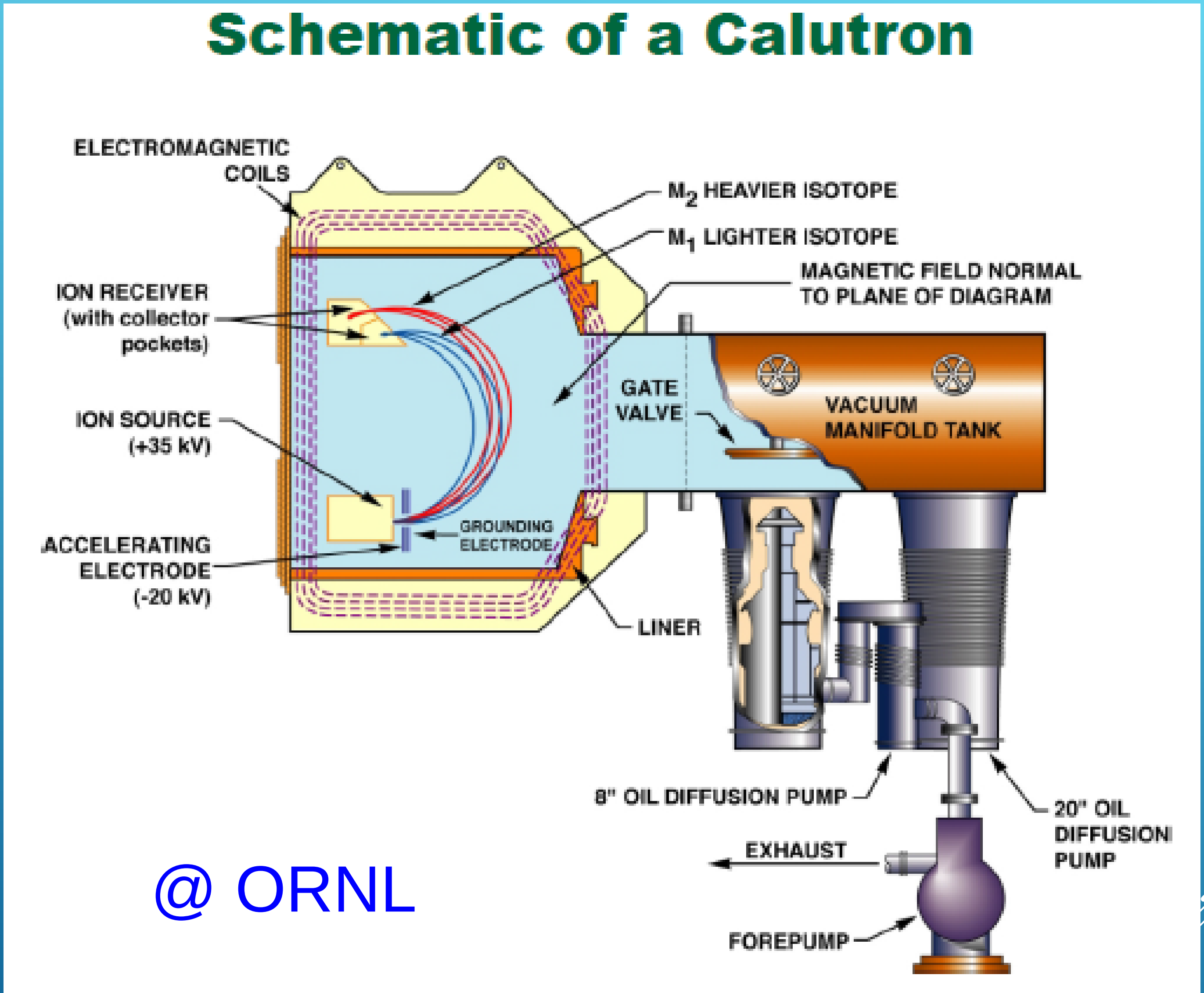
Cherenkov has a faster peak than scintillation.

Natural radiative U/Th decay chain



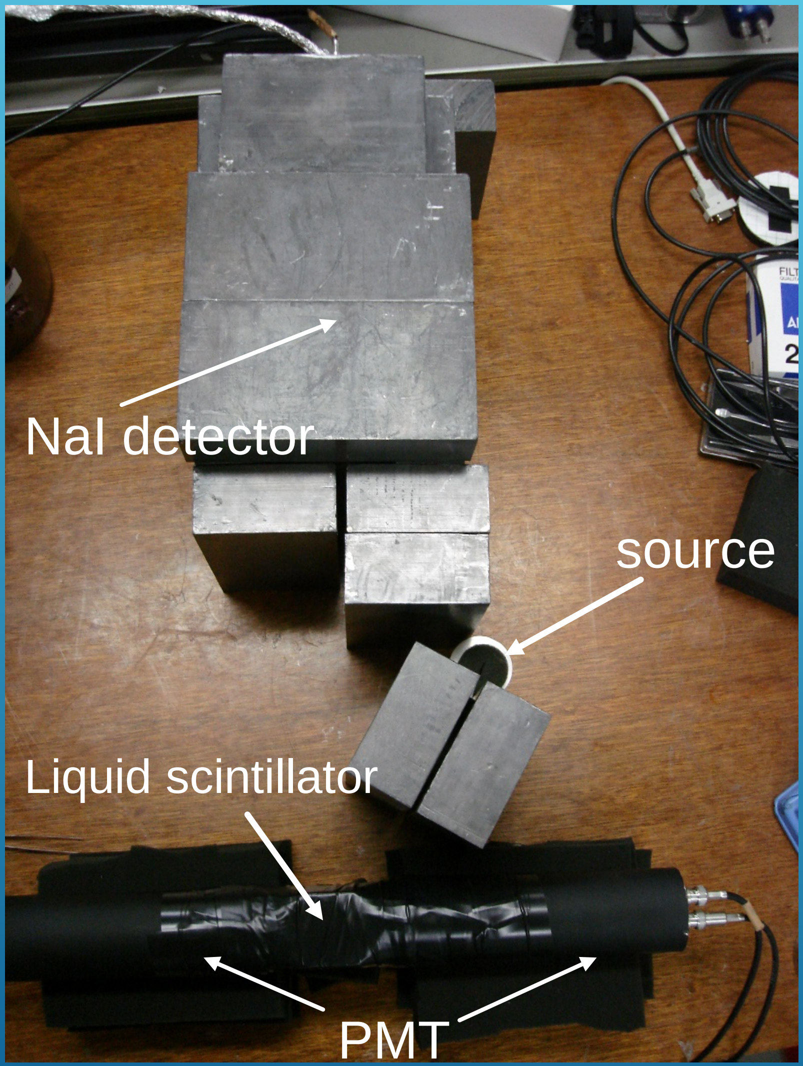


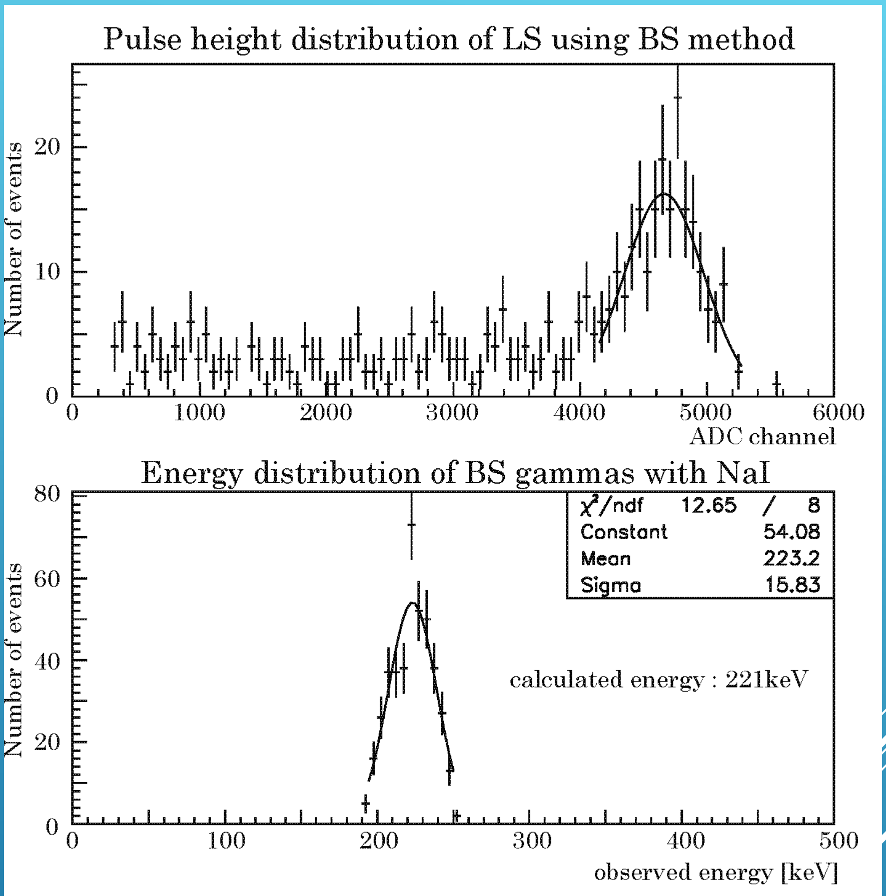
Electromagnetic isotope separation



@ ORNL

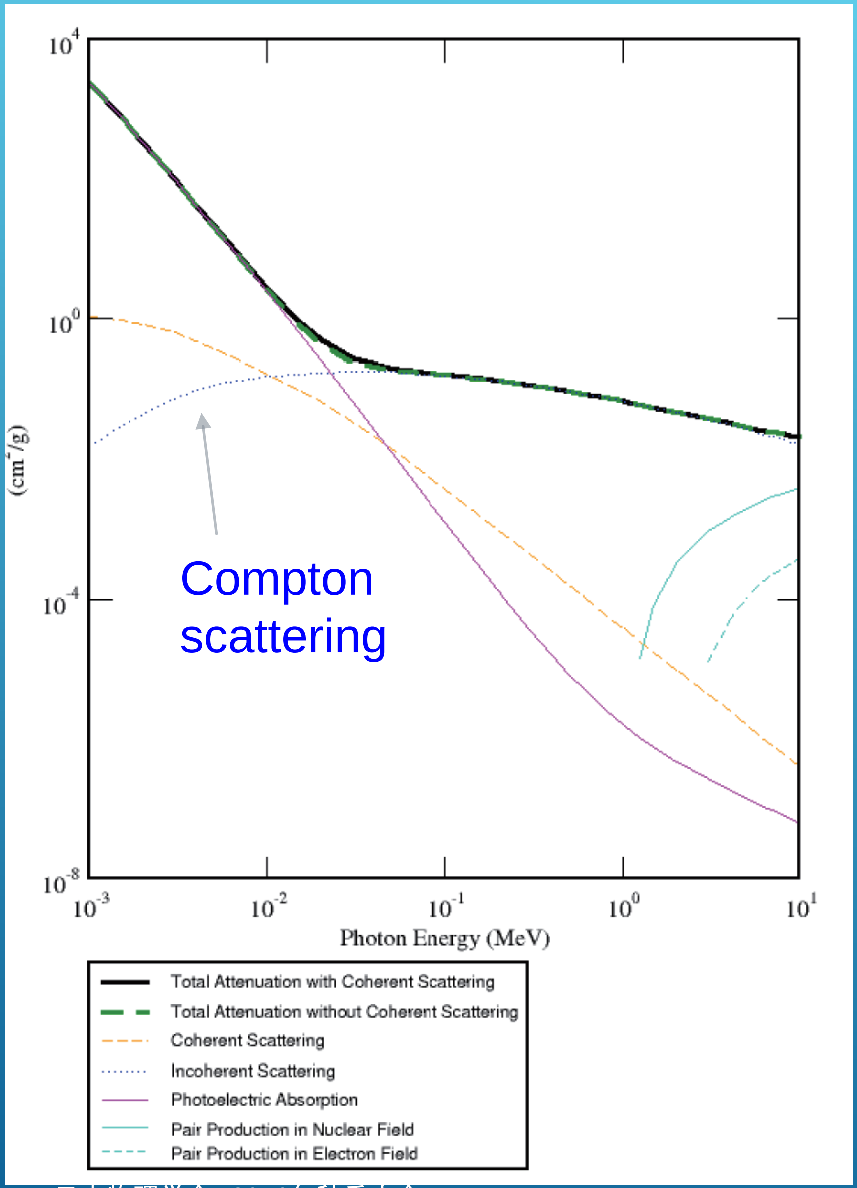
Backscattering method





Single peak could be used even in liquid scintillator.

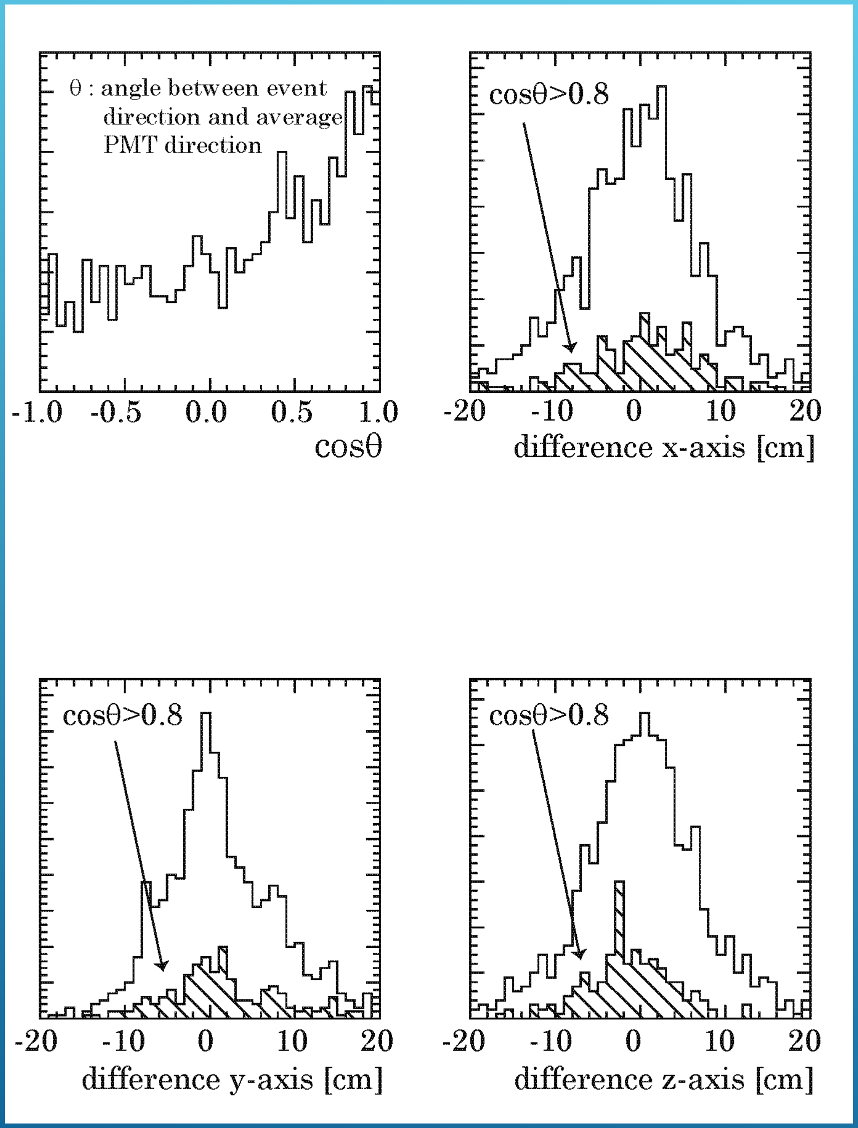
Cross section of gamma in anisole



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

At least, one event could have Cherenkov hits.

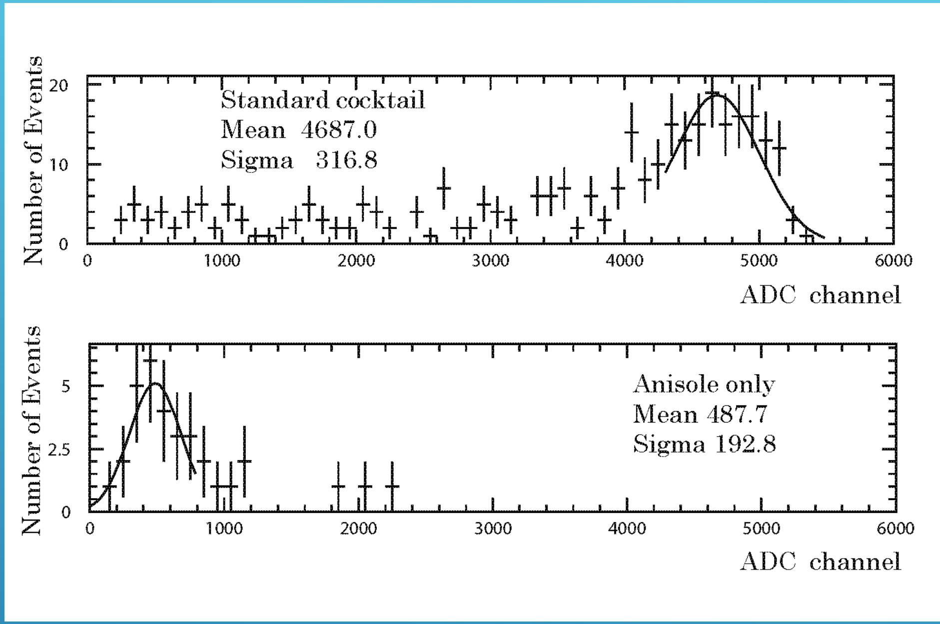
Event direction and average PMT direction

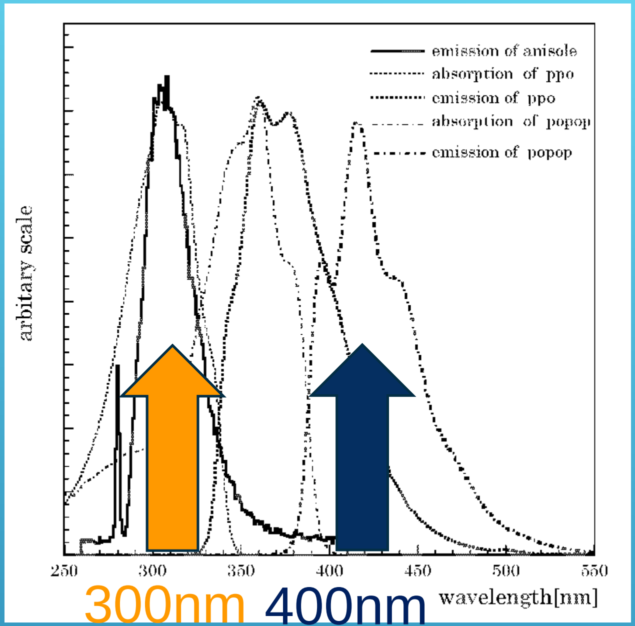


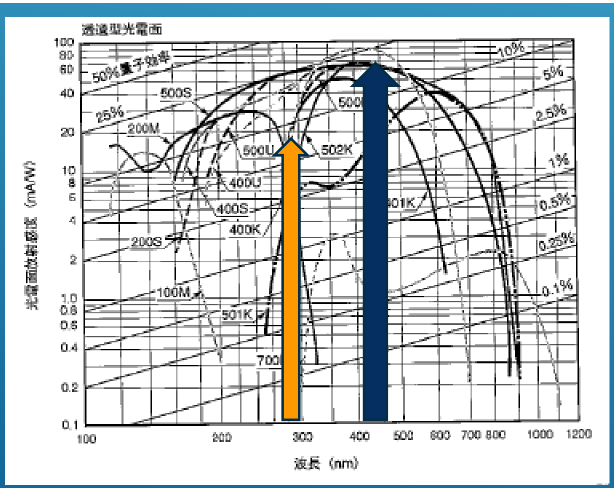
No relation between event direction and averaged PMT direction was found .

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

Light yield of scintillation in anisole

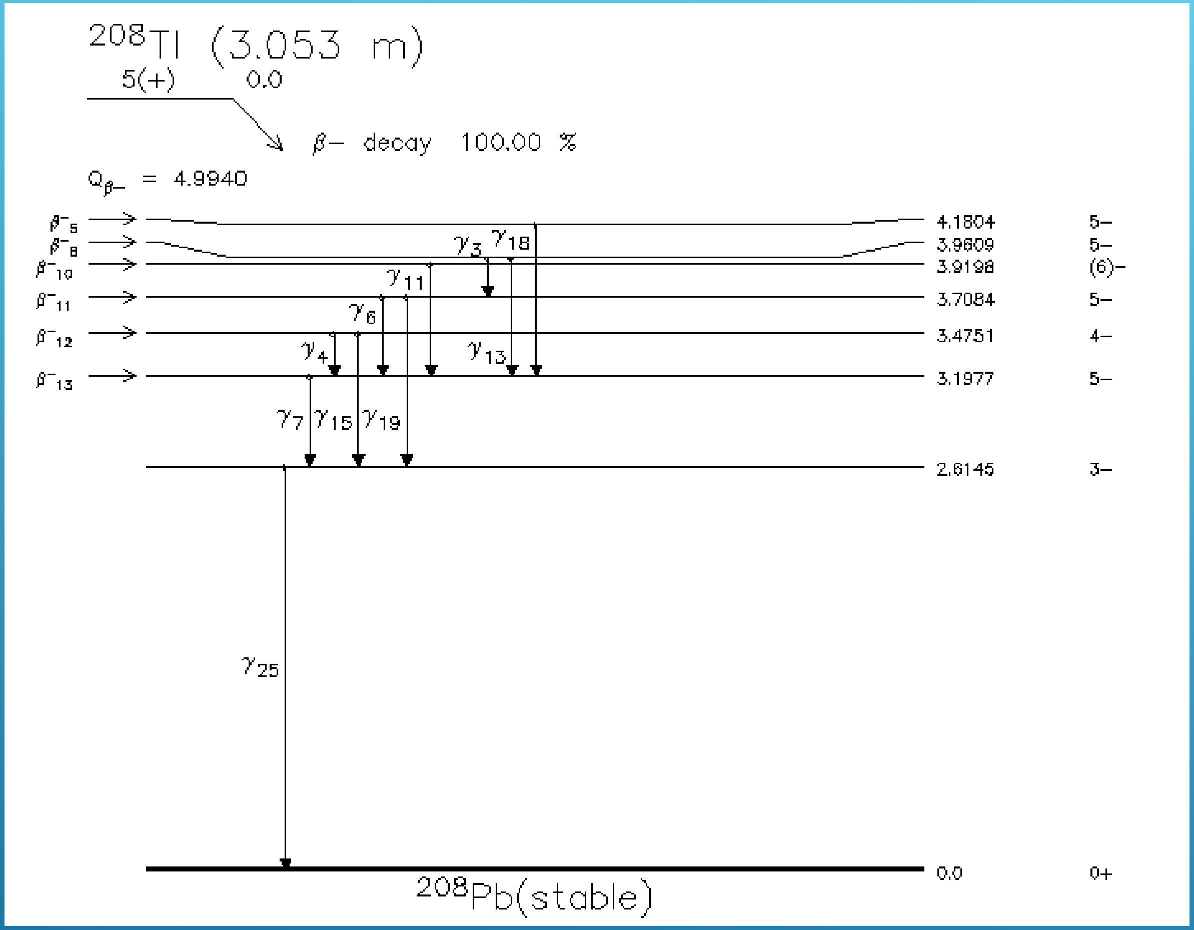






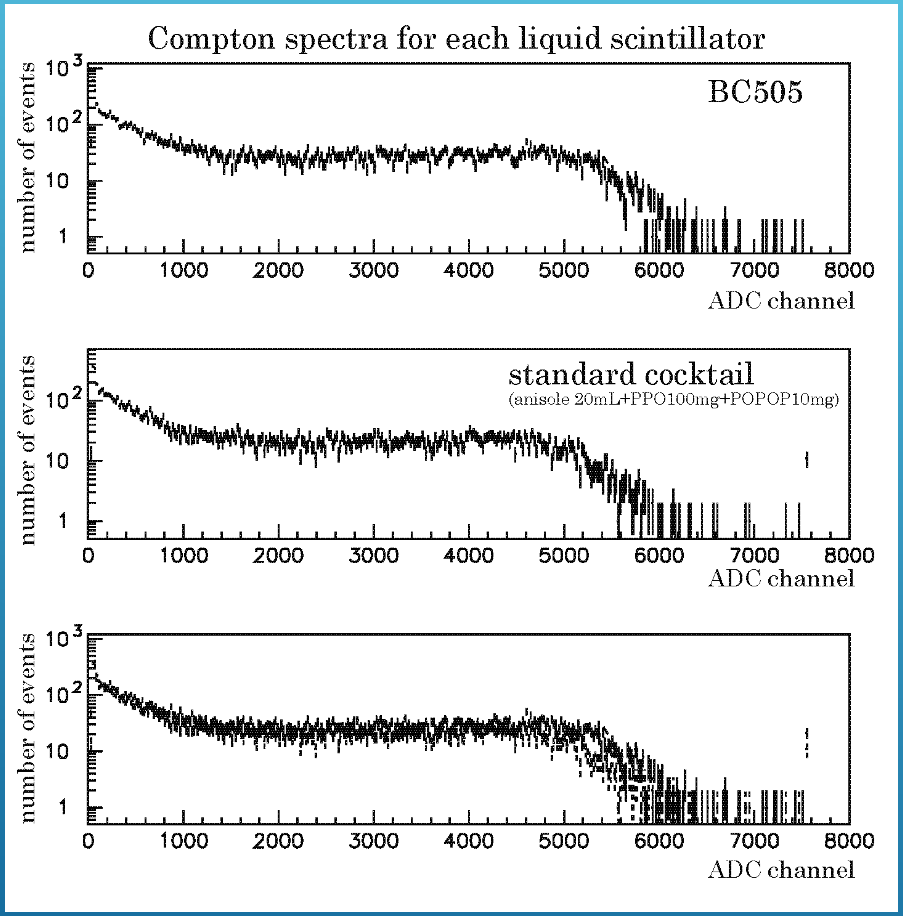
Relative scintillation light yield of anisole is 9.8% to standard cocktail (due to difference of quantum efficiency of PMT)

Thallium-208 radiation branch



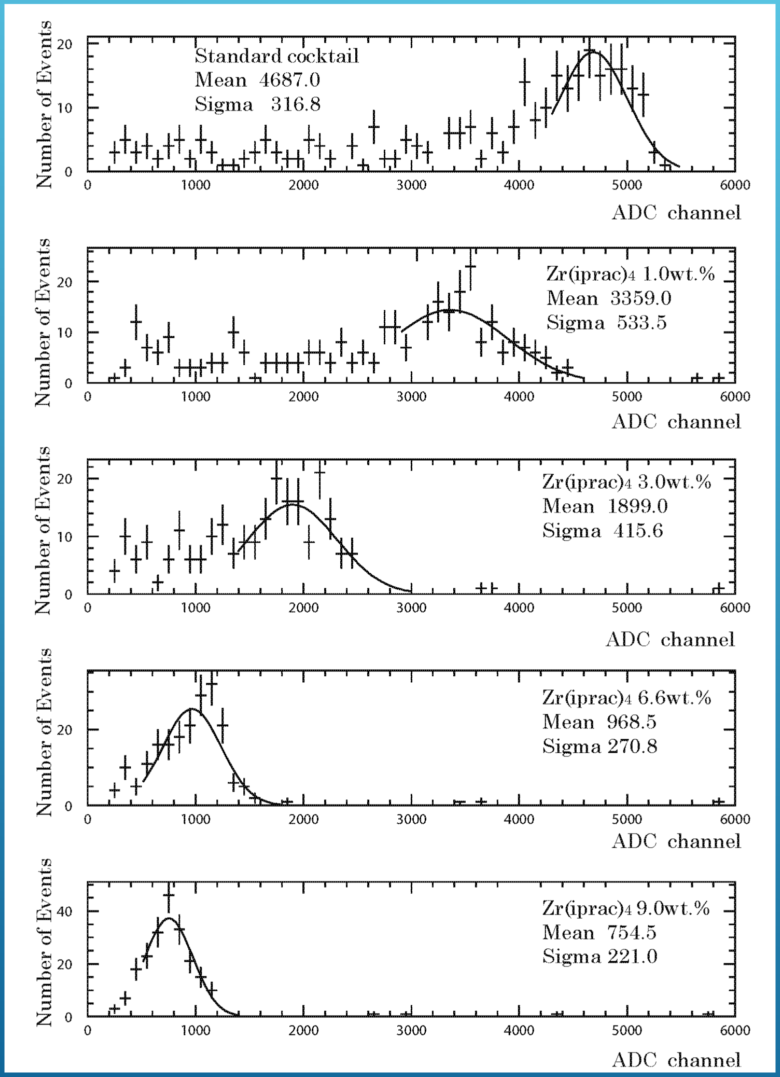
Radiations	y(i) $(Bq\text{-}s)^{-1}$
beta- 5	2.27×10^{-03}
beta- 8	3.09×10^{-02}
beta- 10	6.30×10^{-03}
beta- 11	2.45×10^{-01}
beta- 12	2.18×10^{-01}
beta- 13	4.87×10^{-01}
ce-K, gamma 3	4.04×10^{-03}
gamma 4	6.31×10^{-02}
ce-K, gamma 4	2.84×10^{-02}
ce-L, gamma 4	4.87×10^{-03}
gamma 6	2.26×10^{-01}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

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

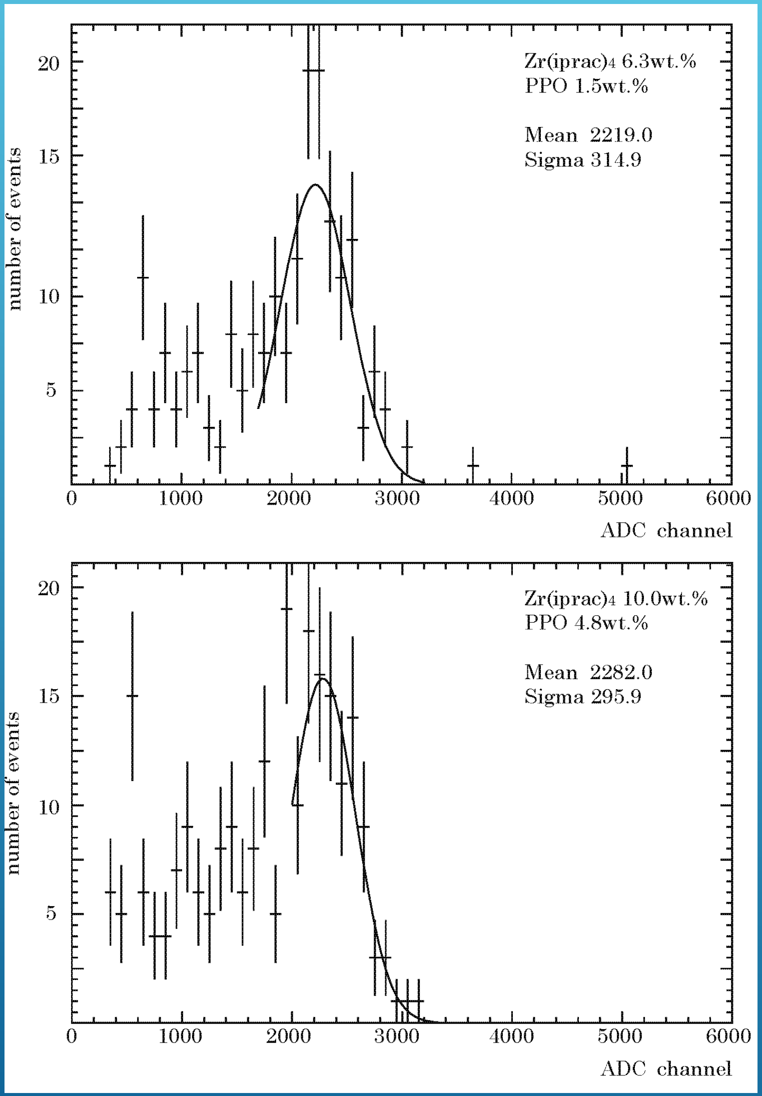
Physical Constants of SGC Liquid Scintillators

Scintillator	Light Output % Anthracene¹	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

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)

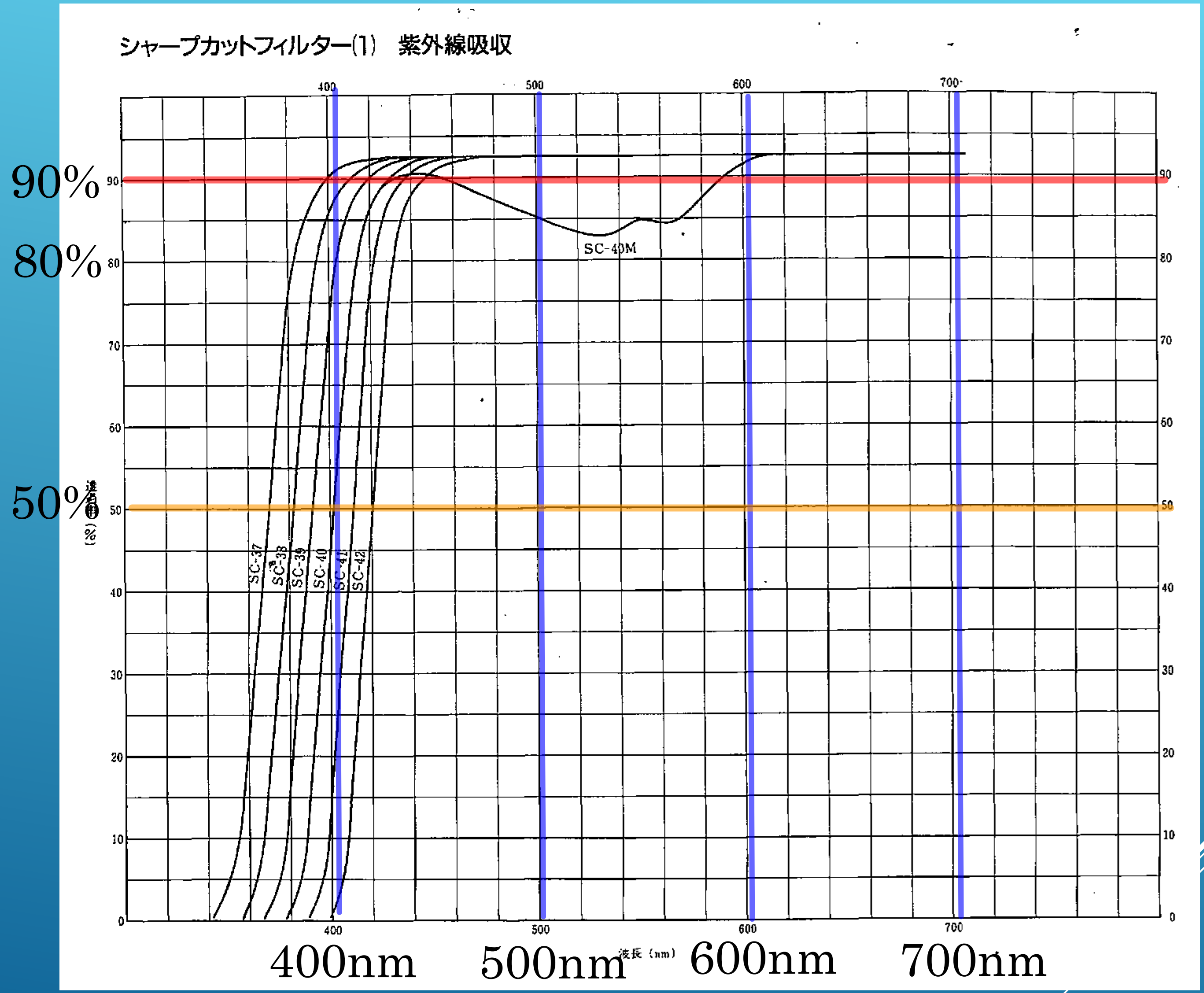
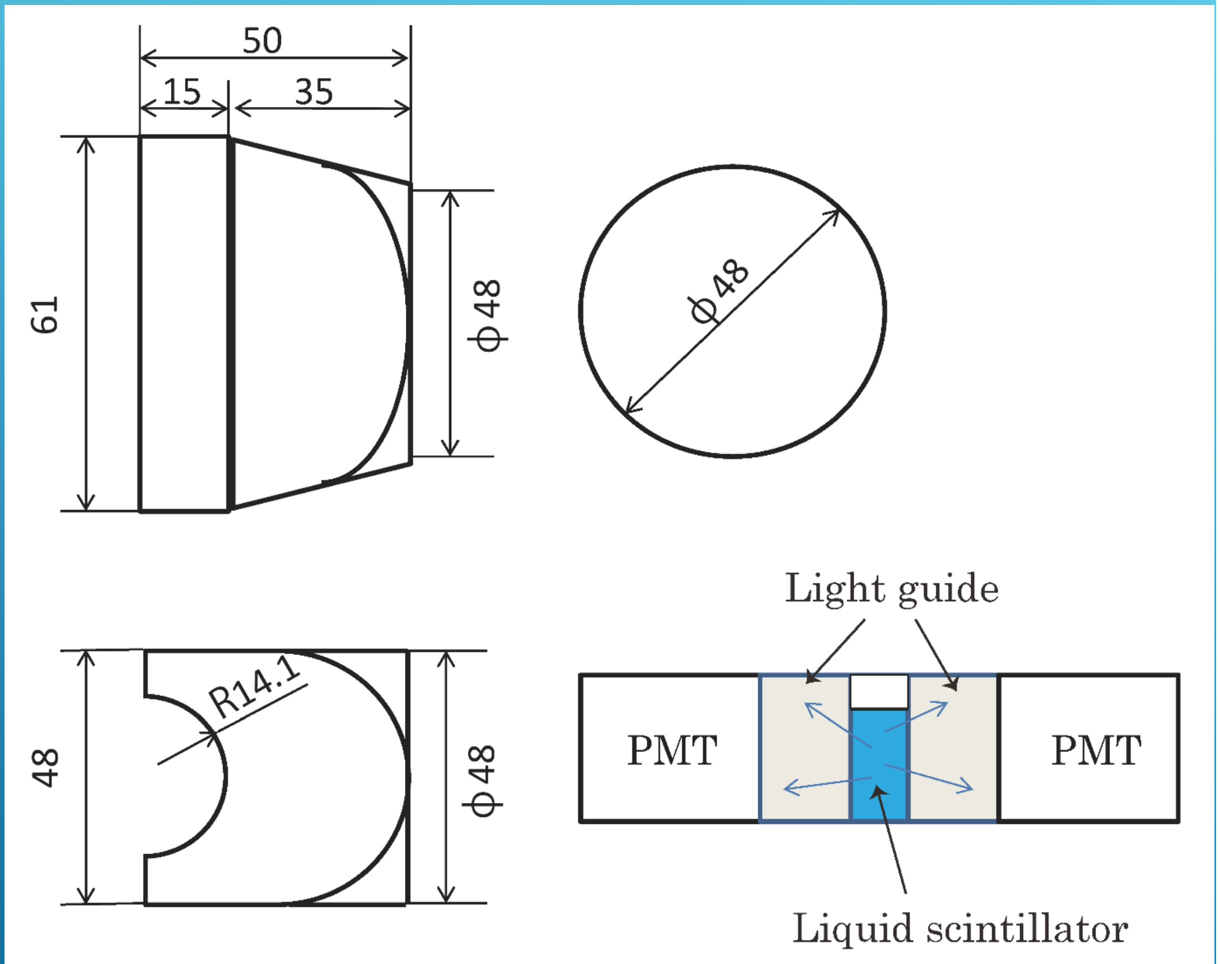
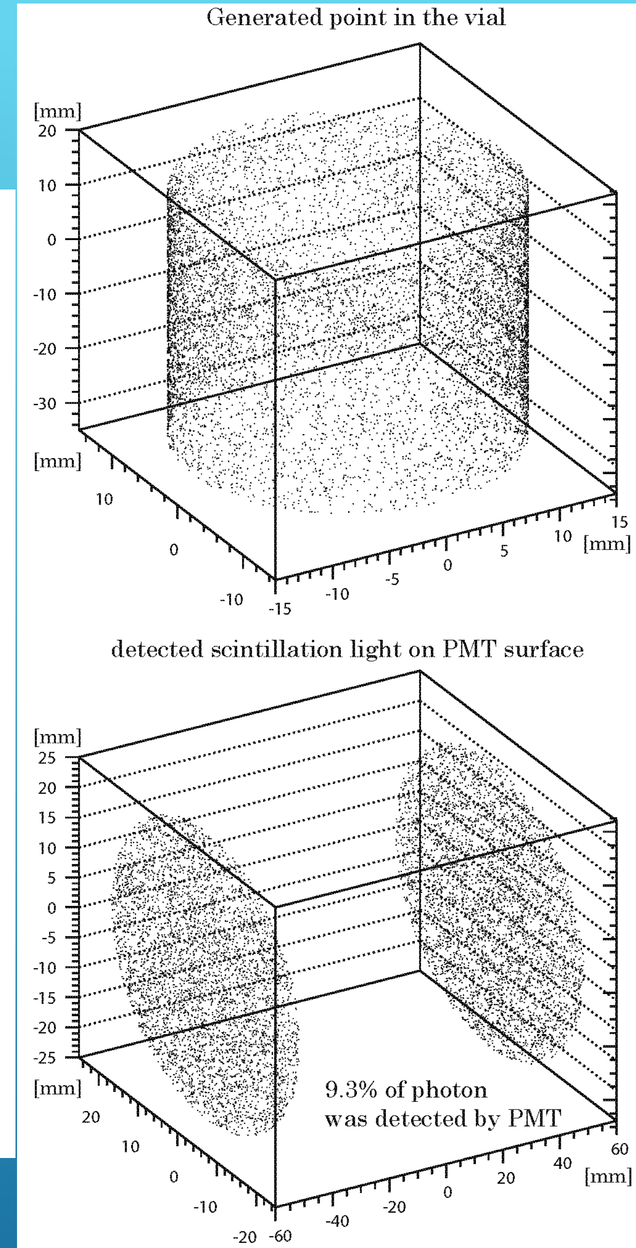
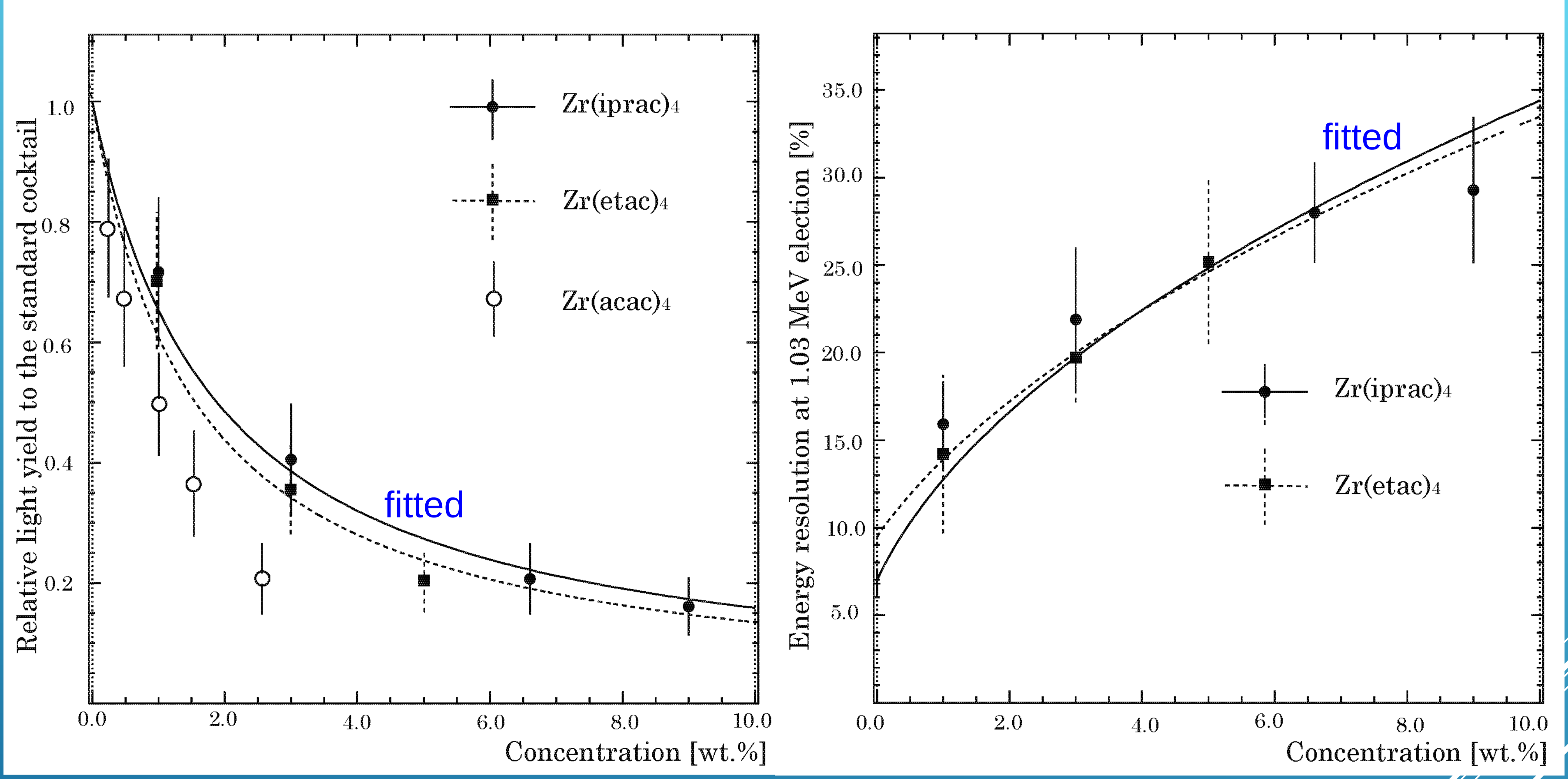


Photo coverage



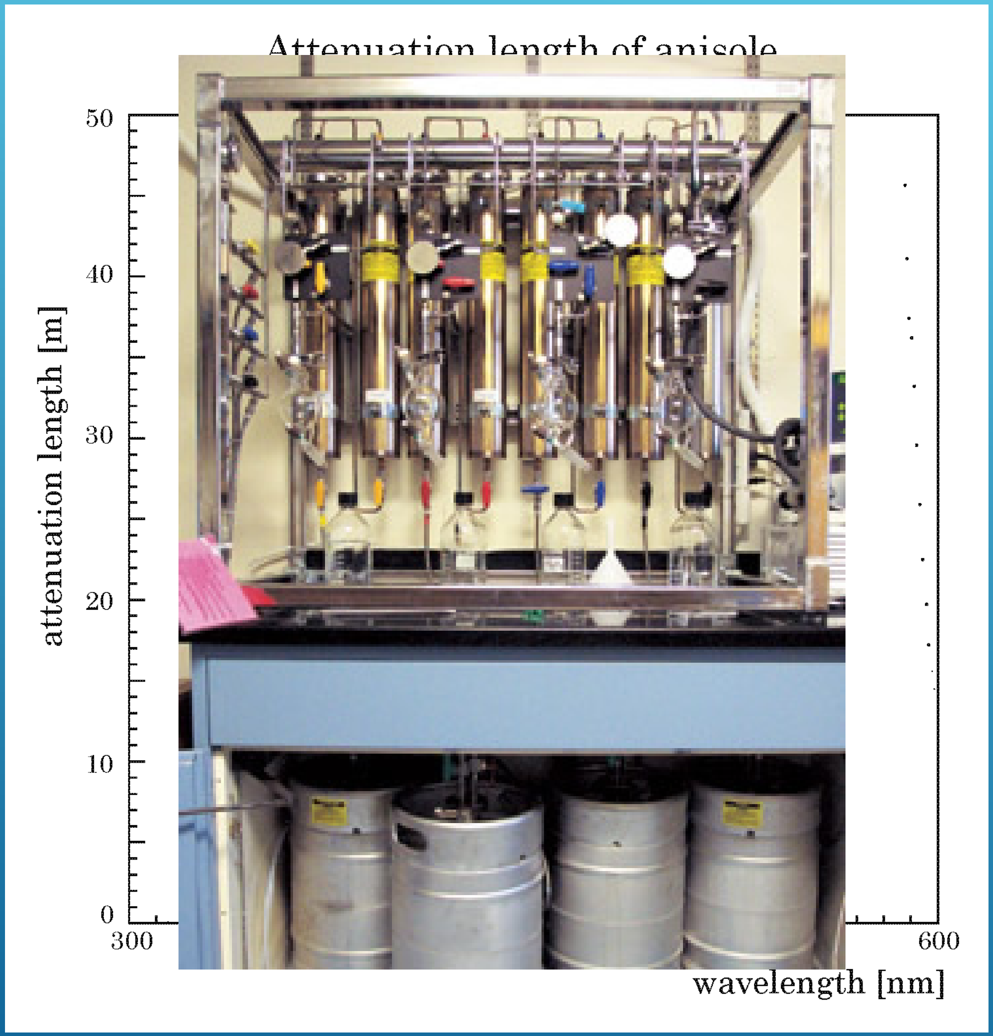


Light yield and energy resolution



$Zr(ipcac)_4$ and $Zr(etac)_4$ have almost same performance, but LY and Eres depend strongly on concemtration.

PURIFICATION OF ANISOLE

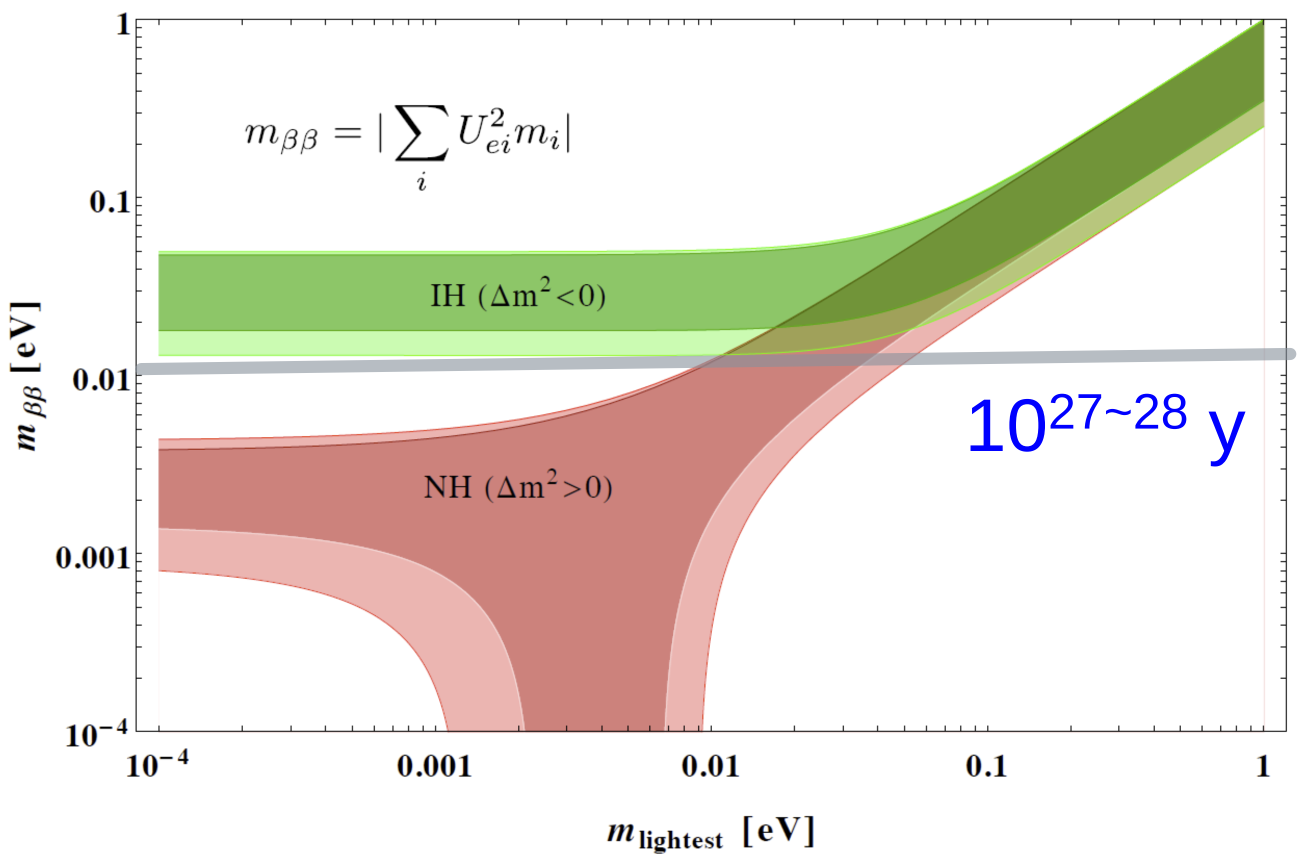


Attenuation length of light from POPOP was obtained as ~6m for current liquid scintillator.

↓

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)

Future 0νββ experiments



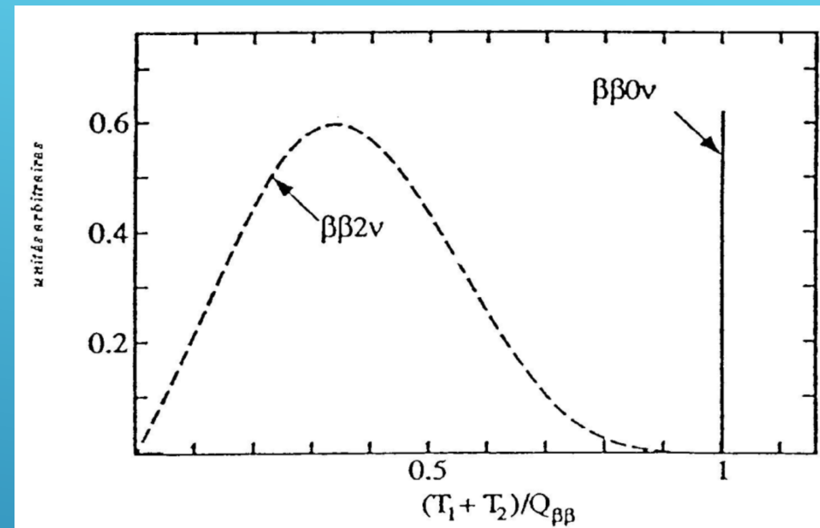
S. Dell'Oro, S. Marcocci, F. Vissani, Phys. Rev. D90, 033005 (2014)

~tons of target and ~zero BG detector will be necessary for next generation 0νββ 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 energy resolution

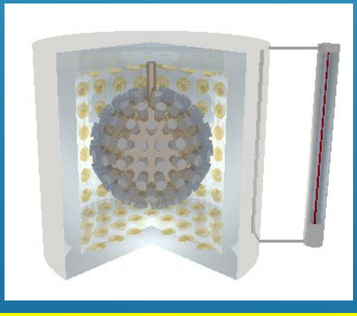
Neutrino mass sensitivity of ZICOS experiment

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

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

Assuming same energy resolution, BG rate and measurement time as KamLAND-Zen ($T_{1/2}^{0\nu} > 2.6 \times 10^{25}$y) (Ref: I.Shimizu arXiv:1409.0077 (2014))

Mass : 113 ton → 10wt.% $Zr(iprac)_4$ = 12.6ton includes 1.7ton of Zirconium = 45 kg of ^{96}Zr **(natural abundance 2.6%)**
(64kg of ^{136}Xe = 0.2 × KL-Zen)



$T_{1/2}^{0\nu} > 1.2 \times 10^{25}$y ← Not enough for $0\nu\beta\beta$ search

Neutrino mass sensitivity of ZICOS experiment

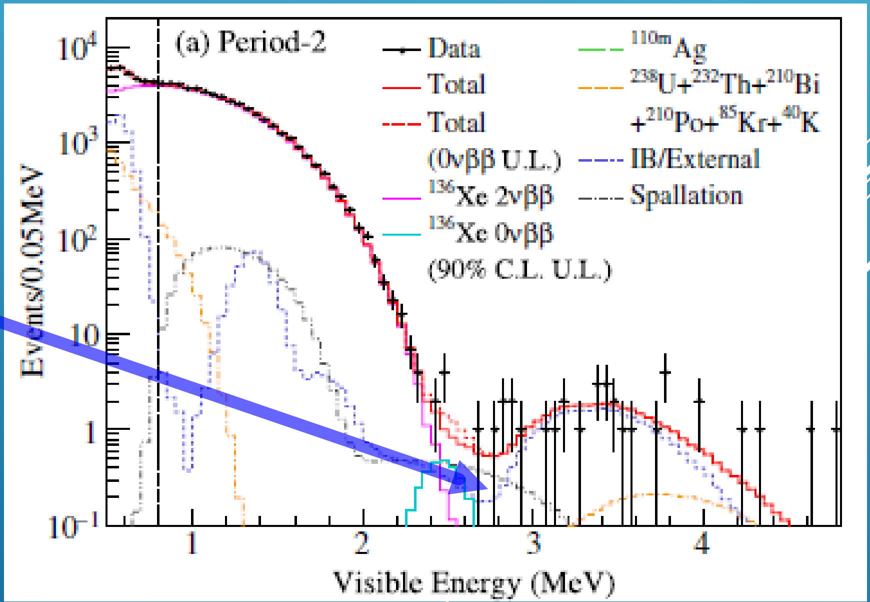
1) Zr enrichment

58.5% enrichment of ^{96}Zr (e.g. 57.3% for NEMO-3)
then ^{96}Zr will be 1.0 ton (4.4 times ^{136}Xe 320kg)

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

Laser

2) Lowering BG (^{208}Tl /^{214}Bi)
i.e. < 1/20 × KL-Zen
(~1.0events/ton/year)

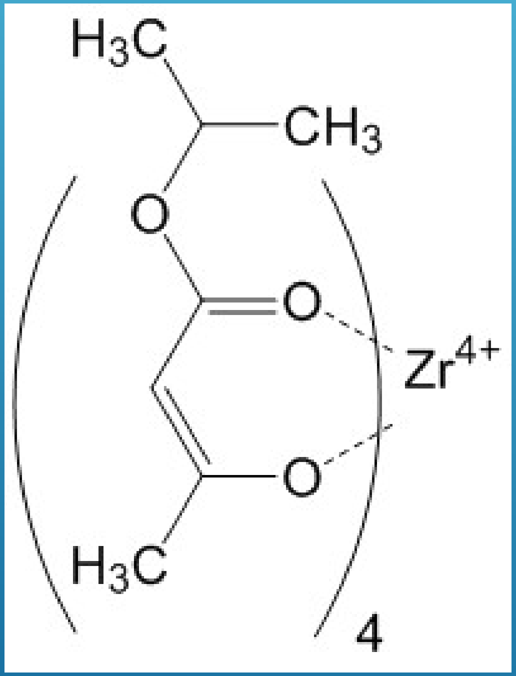


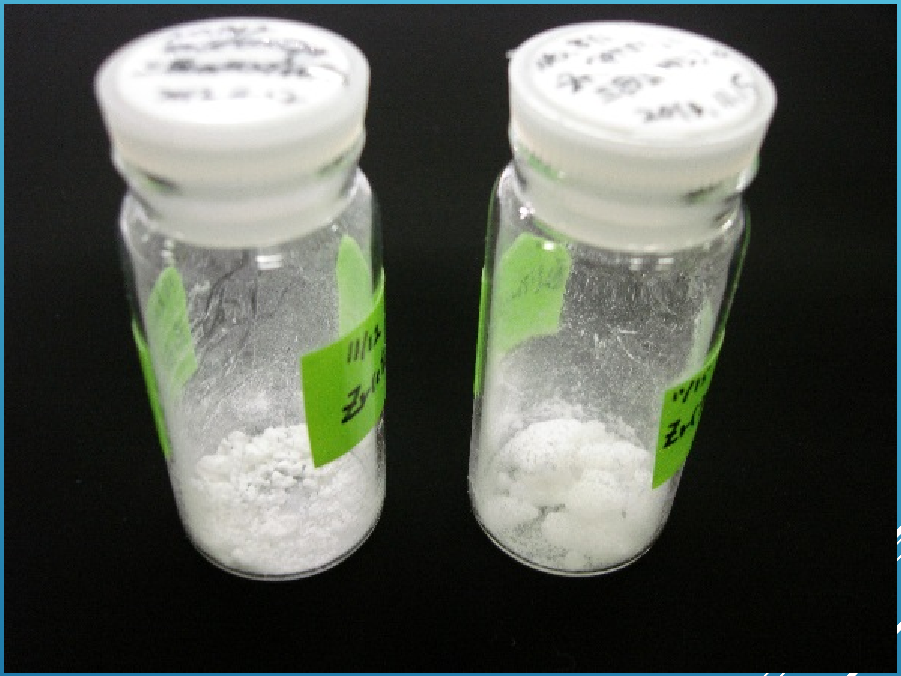
$T_{1/2}^{0\nu} > 5 \times 10^{25}$y

See this talk

Development of Zr loaded LS

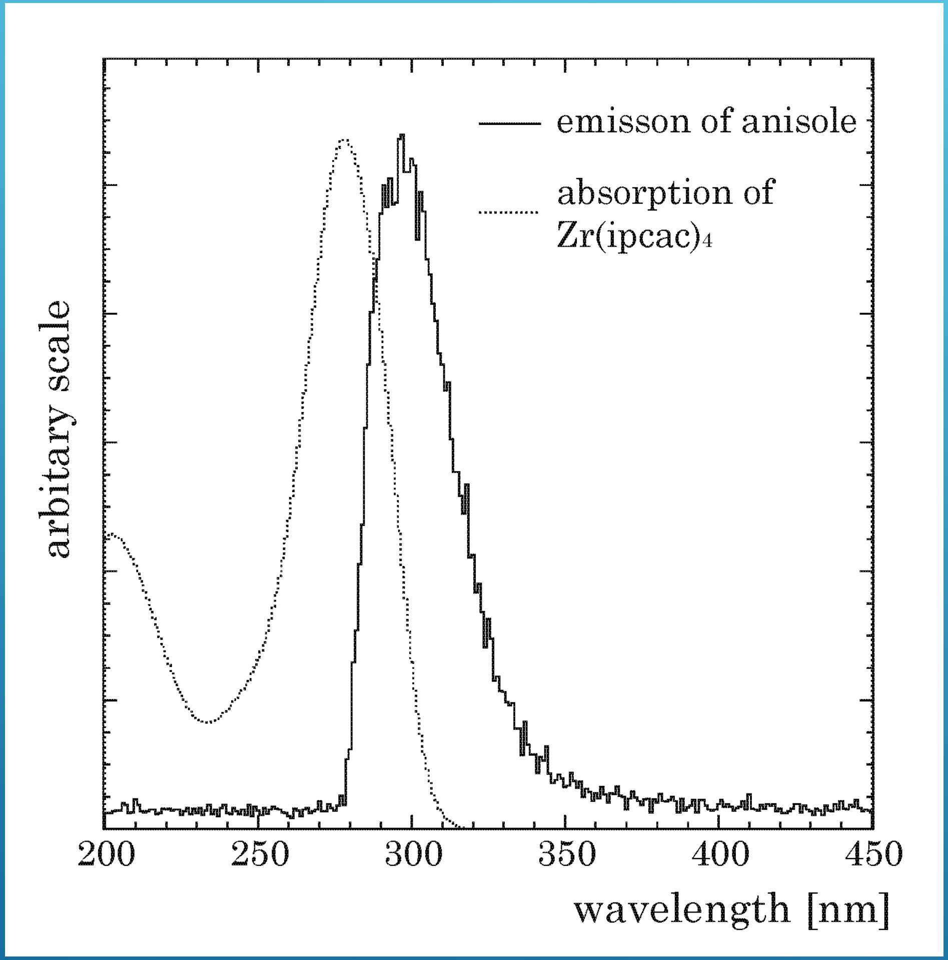
$Zr(CH_3COCHCOOCH(CH_3)_2)_4$: $Zr(iprac)_4$
tetrakis (isopropyl acetoacetate) zirconium
mw : 663.87





Solid crystal or powder

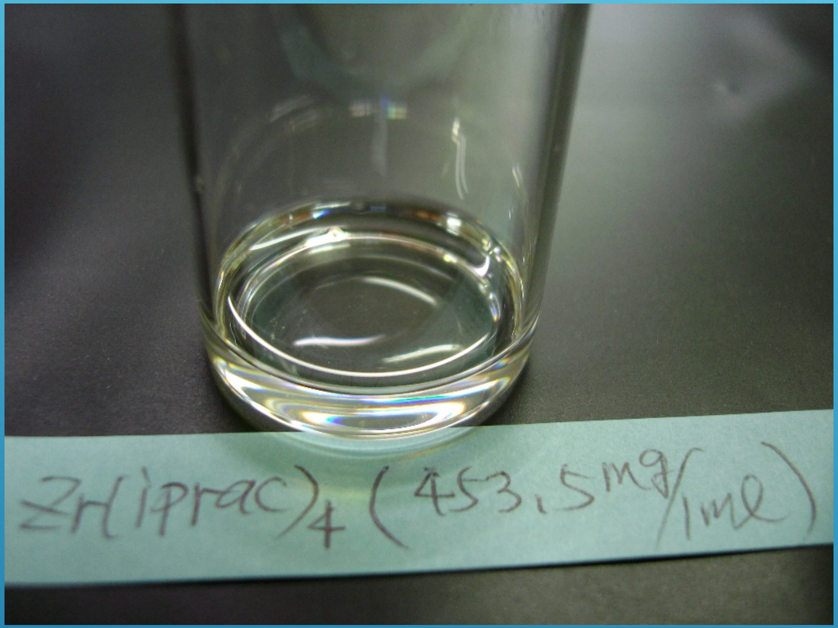
Absorbance spectra for Zr(iprac)$_4$

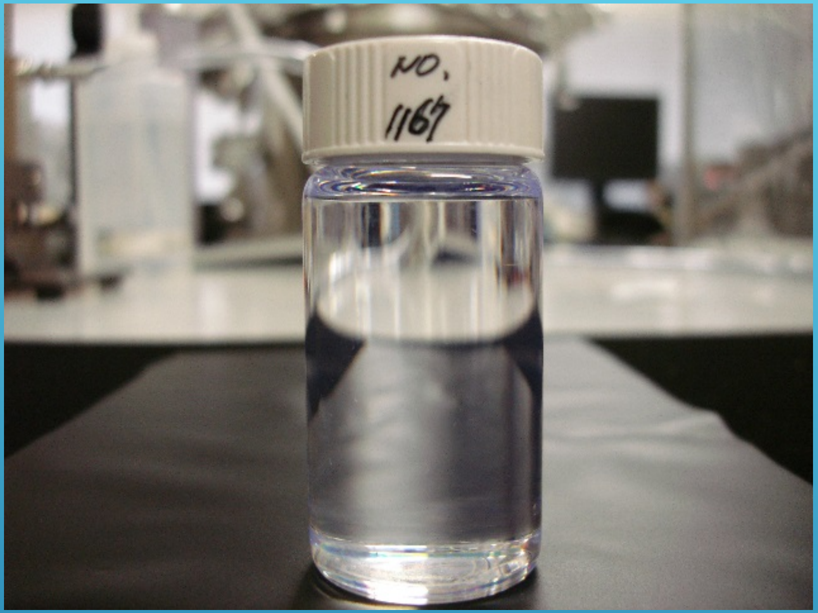


Absorption peaks of Zr(iprac)$_4$ was found around at 278nm. However, overlapped region with emission of anisole was existed.

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 $Zr(iprac)_4$

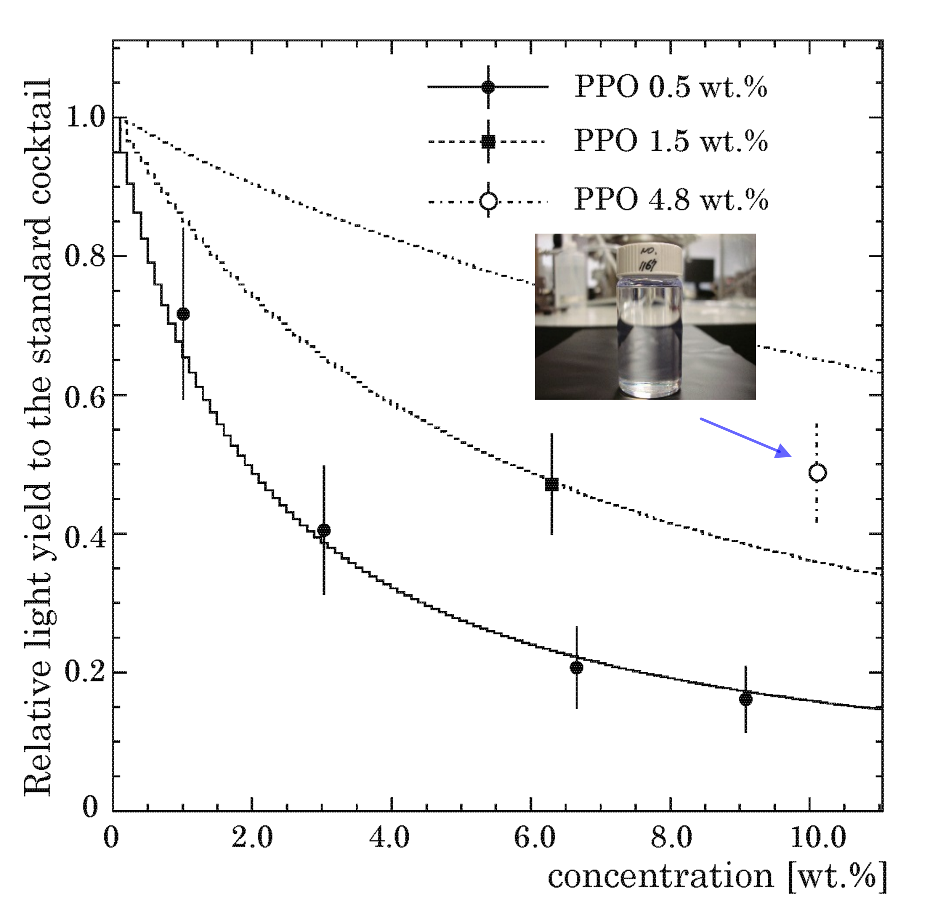
$$\text{Light yield} = L_0 \times \frac{\sigma_1 N_{ppo}}{\sigma_1 N_{ppo} + \sigma_2 N_{Zr}}$$

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

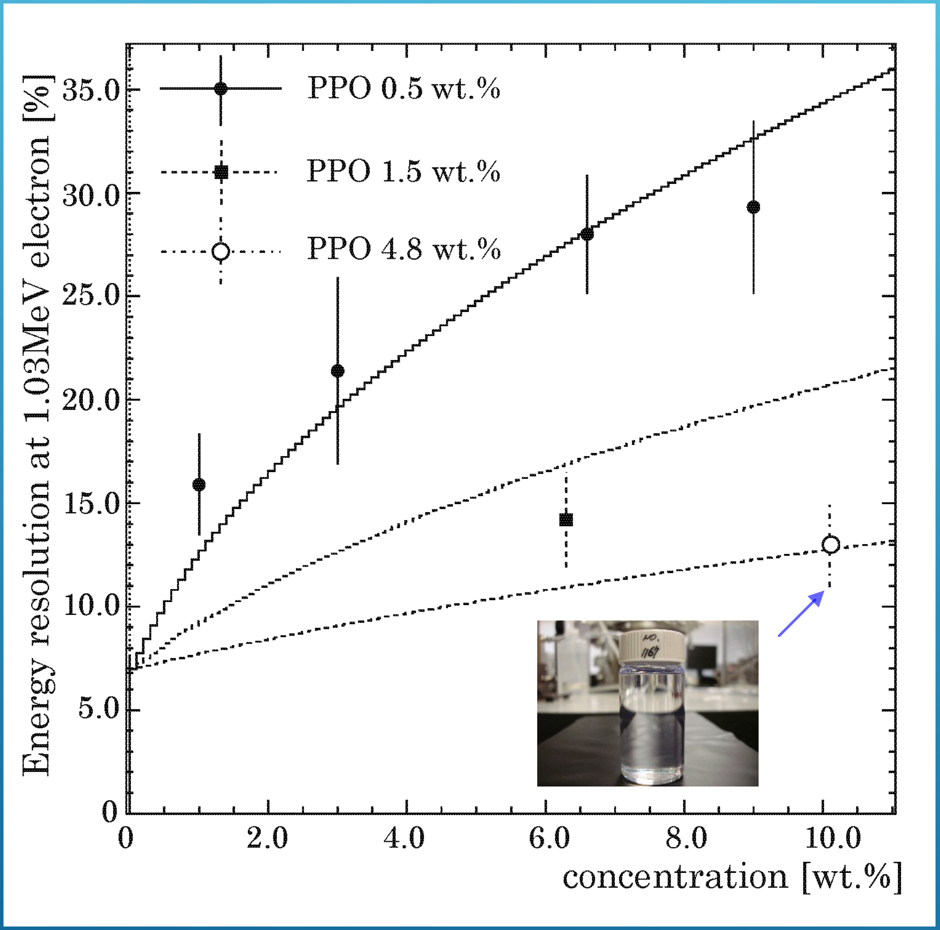


5wt.% PPO helps actually recovering the scintillation light yield.

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 $Zr(iprac)_4$.

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

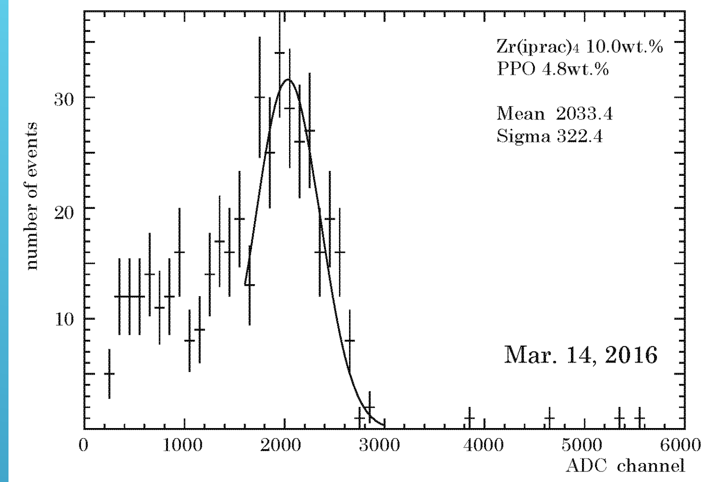
= 3.5±0.5% at 3.35MeV

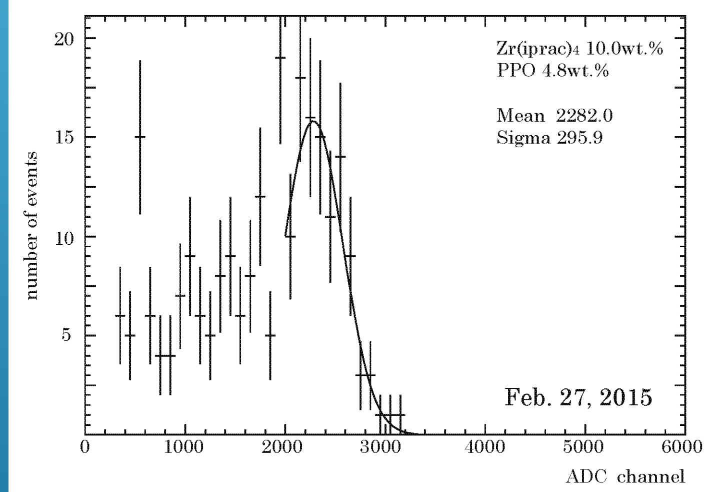
Achieved goal !

Stability of liquid scintillator

Feb. 27,2015

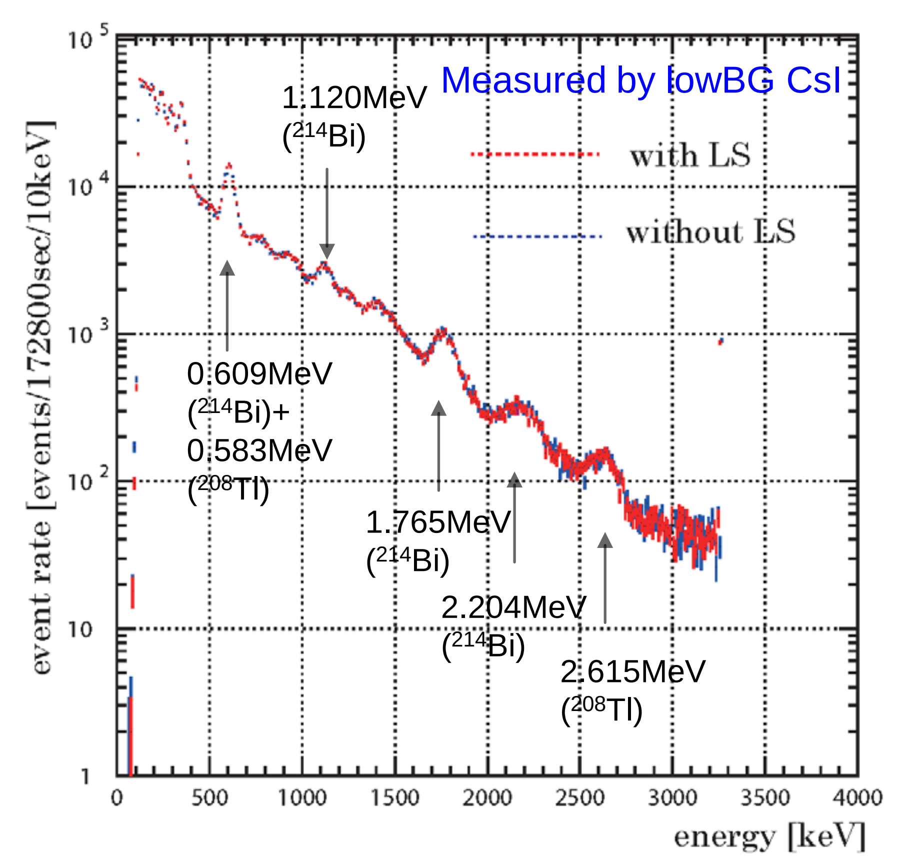
Mar. 14, 2016





Keep transparent liquid and no precipitate is found.

Measurement of backgrounds from LS



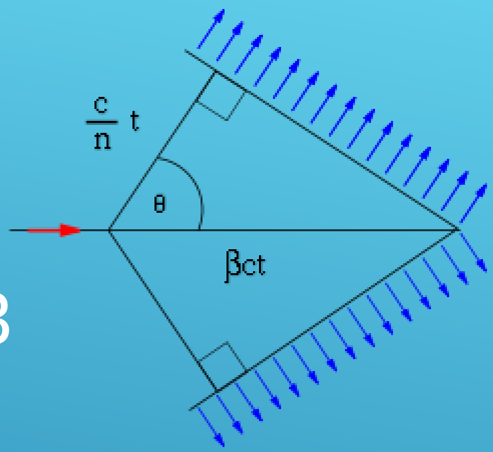
Using subtracted # of events around 2.6MeV and 2.2MeV

^{214}Bi < 4.9x10^{-20}g/g ^{208}Tl < 2.7x10^{-22} g/g

(^{238}U < 6.4x10^{-6} g/g) (^{232}Th < 7.4x10^{-7} g/g) (c.f. KL 10^{-18}g/g)

Property of Cherenkov light

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

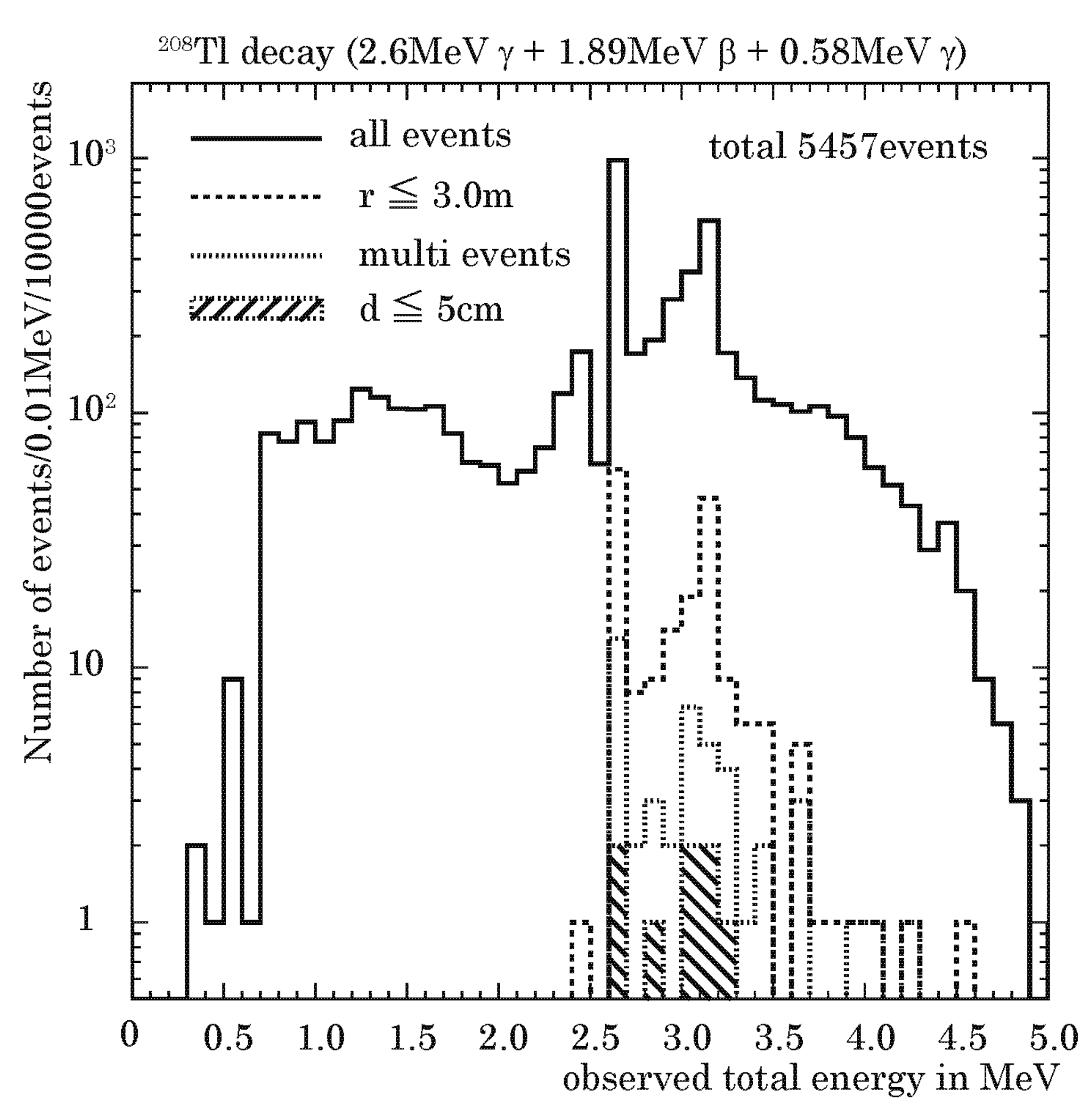


$$\frac{dN}{dx} = 2\pi z^2 \alpha \sin^2\theta_c \int_{\lambda_1}^{\lambda_2} \frac{d\lambda}{\lambda} = 475 z^2 \sin^2\theta_c \mathrm{photon/cm}$$

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

Cherenkov light = ~1% of scintillation light

Simulation of ^{208}Tl decay



1) E : 3.0-3.7MeV
   1555 events
2) fiducial volume
   91 events
3) Multi events
   22 events
4) Closer events
   5events

1/20 BG reduction could be achieved