

DEMONSTRATION OF ZICOS EXPERIMENT FOR OBSERVATION OF ZIRCONIUM-96 TWO NEUTRINO EMISSION DOUBLE BETA DECAY

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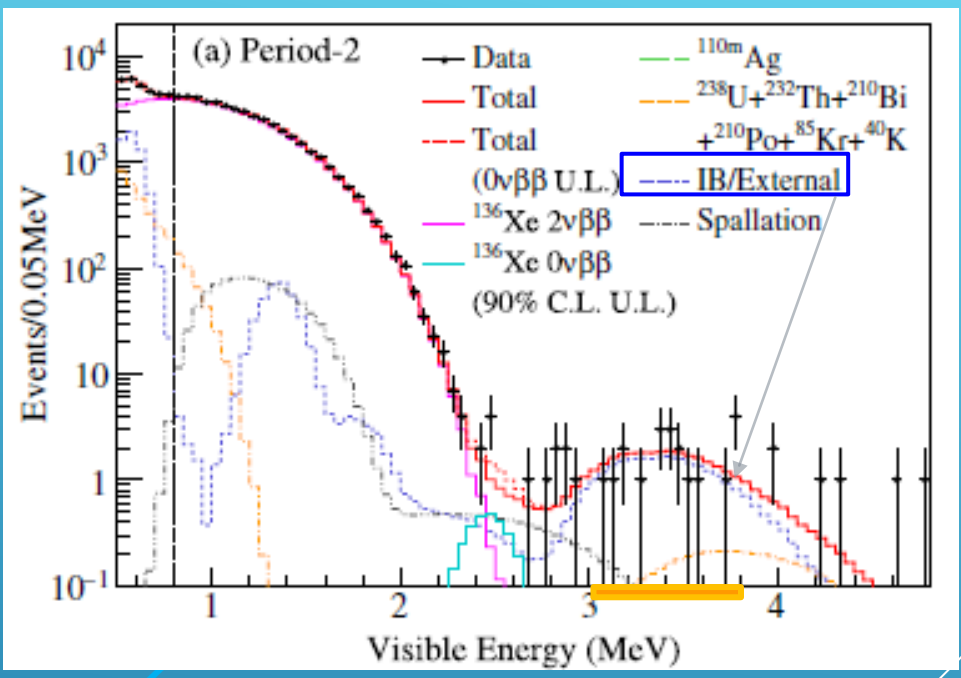
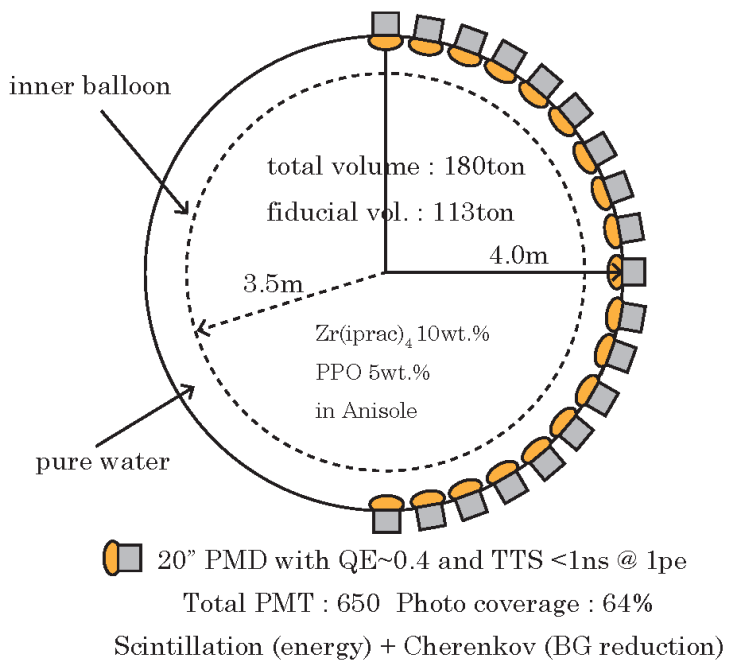
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Conceptual design of ZICOS detector

Phys.Rev.Lett. 117 (2016) 082503

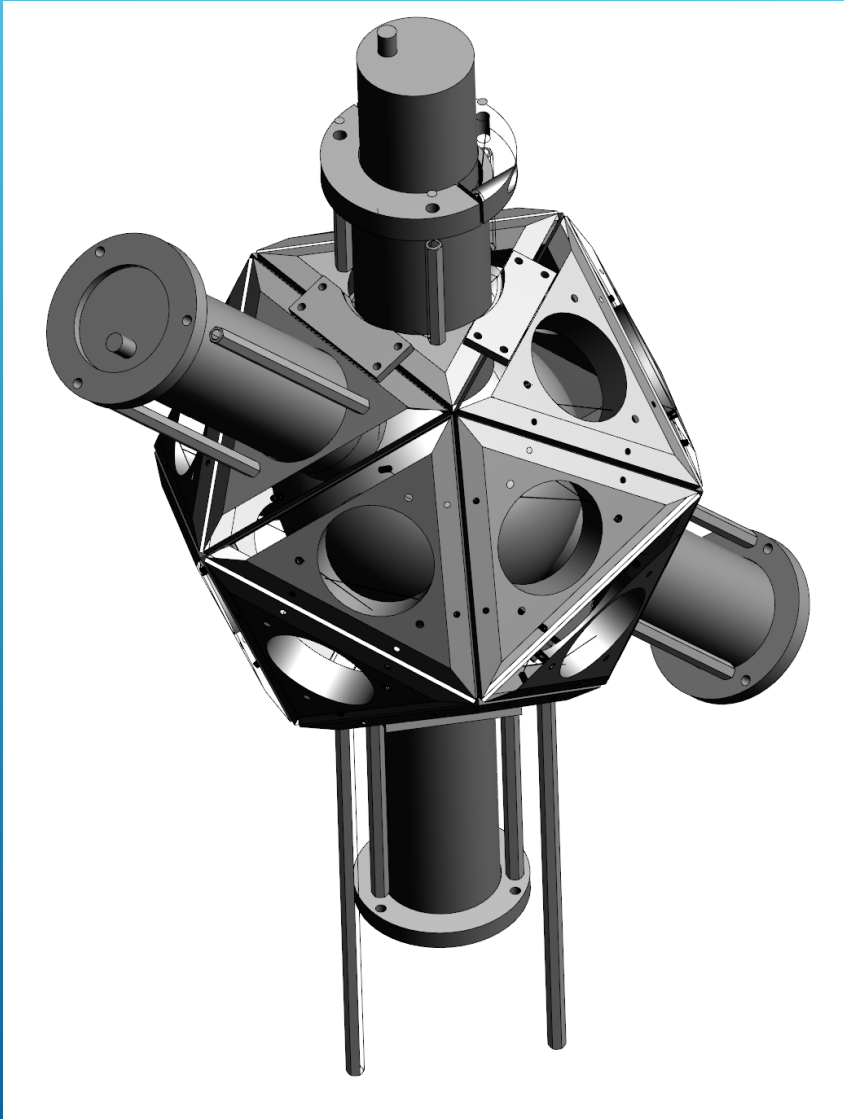
Conceptual design of ZICOS detector



NEMO3 : $T_{1/2}^{0\nu} > 9.1 \times 10^{21}$ yrs

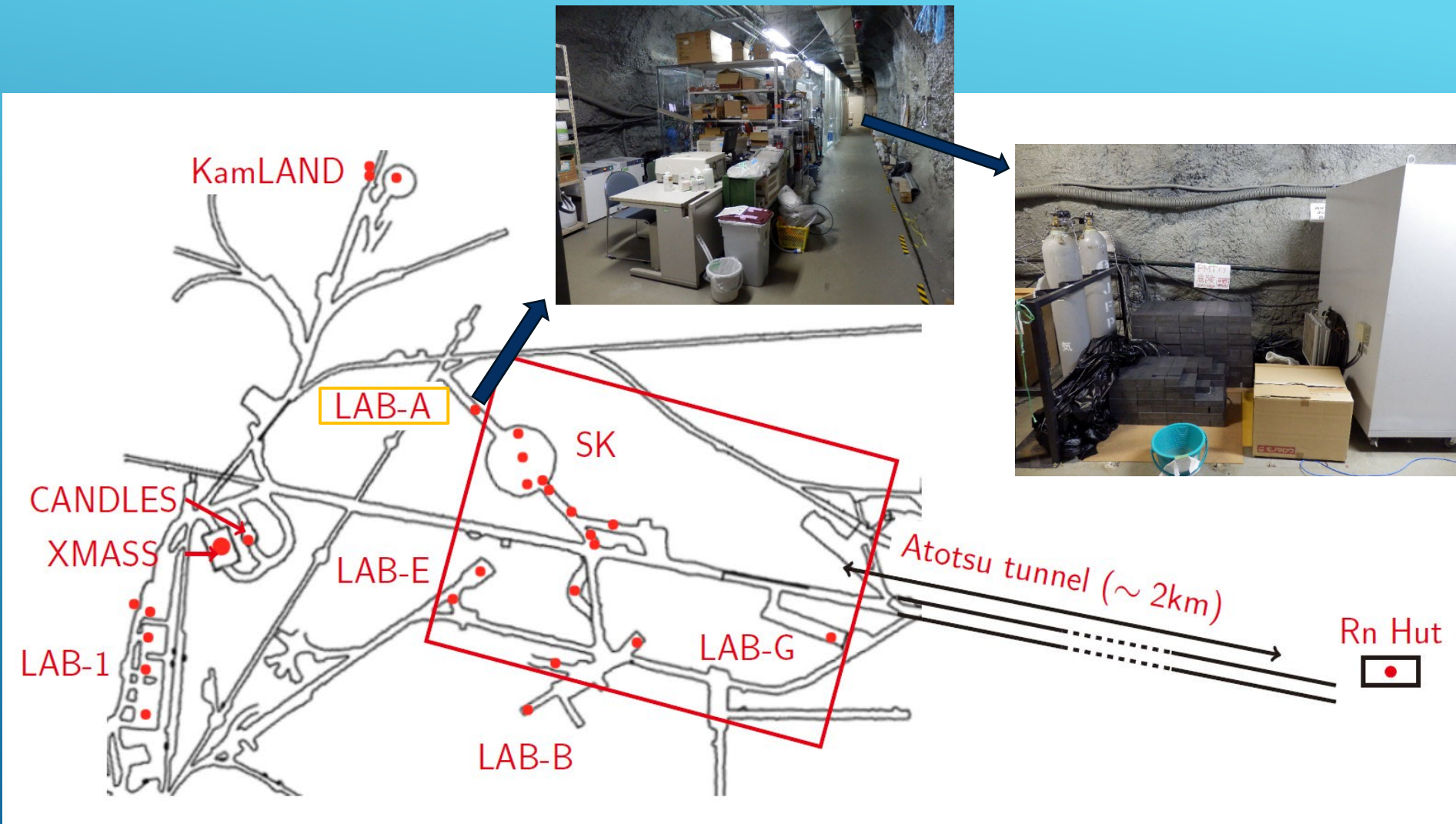
**^{96}Zr : 45 kg (nat.) \rightarrow 865 kg(50 % enrich) \rightarrow 1/20 BG
 $T_{1/2}^{0\nu} > 4 \times 10^{25}$ yrs $\rightarrow 2 \times 10^{26}$ yrs $\rightarrow \sim 1 \times 10^{27}$ yrs**

Observation of $2\nu\beta\beta$ events using ^{96}Zr



- 16 cm diameter round bottom flask using Ultra-pure quartz.
- 20 low BG 2" PMT Hamamatsu H3378-50.
- Designed mounting jig
- Filled 1.0L of ZICOS liquid scintillator loaded 100g of $\text{Zr}(\text{iPrac})_4$ which contains 0.4 g of ^{96}Zr .
- Expected number of events is ~ 100 per year.
- LAB-A in Kamioka mine

Underground laboratory in Kamioka mine



LAB-A : Behind of LINAC control room

Background estimation

U/Th in GE214 using ICP Mass spectrometer :

^{232}Th : 15ng/g corresponds to $6.09 \times 10^{-5}\text{Bq/g}$

^{238}U : 29ng/g corresponds to $3.58 \times 10^{-4}\text{Bq/g}$

^{40}K : 0.021ng/g corresponds to $5.59 \times 10^{-6}\text{Bq/g}$

Assuming radiation (perpetual) equilibrium :

$$\lambda_A N_A = \lambda_B N_B \text{ (Decay rate should be same)}$$

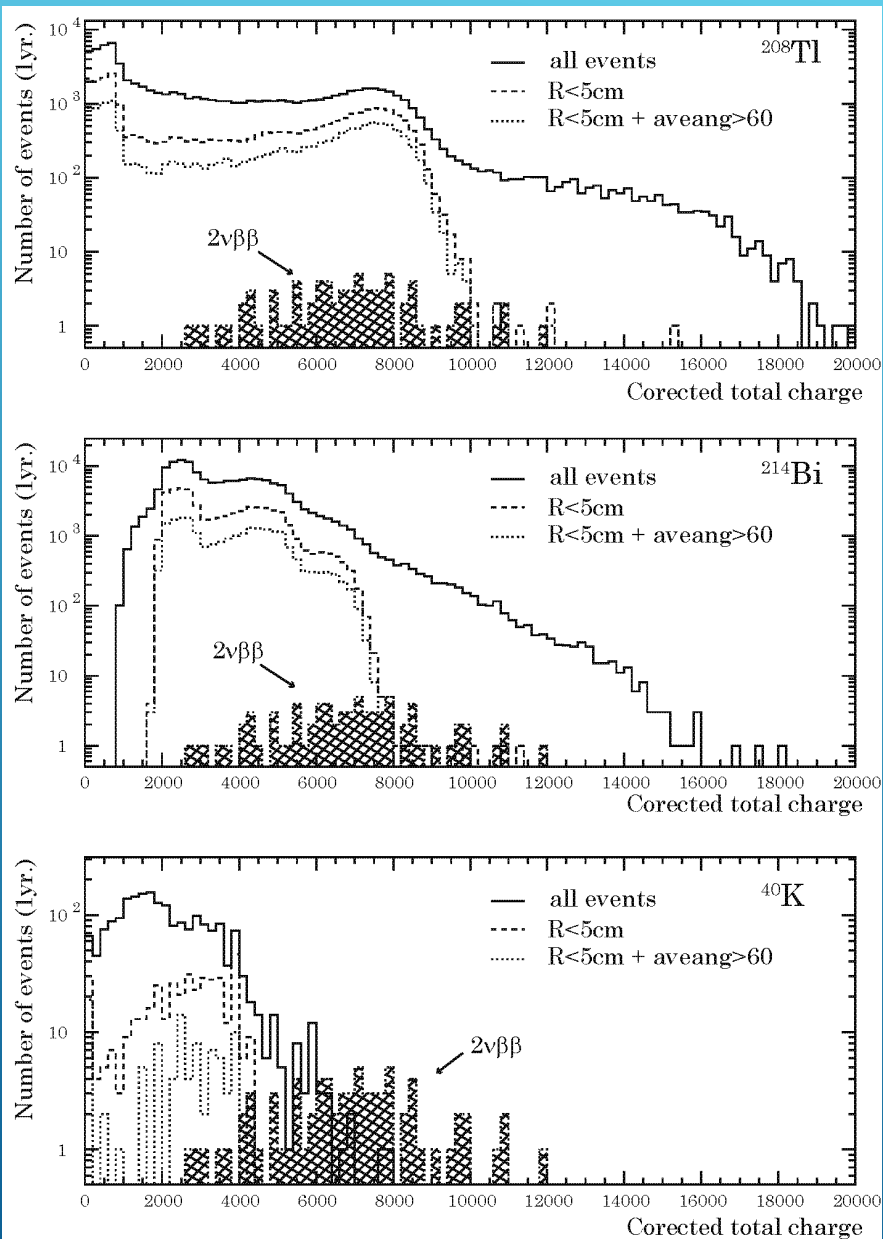
The detector flask uses 530g of GE214.

^{208}Tl : 1017908 events per year

^{214}Bi : 5988404 events per year will occur.

^{40}K : 93556 events per year

Monte Carlo simulation assuming ETFE bag



- ^{40}K affects only part of $2\nu\beta\beta$ observation.
- ^{214}Bi is significant BG, but small fraction of $2\nu\beta\beta$ events should be observed.
- ^{208}Tl is most serious BG for $2\nu\beta\beta$. A few events might be observed.

Need tuning for inner ETFE cubic bag for liquid scintillator.

Present status

- Clean booth (class 1000) was constructed for preparation of Liquid Scintillator and detector.
- 200g of Zr(iPrac)_4 is ready. Stored in the dry box.
- 20 Hamamatsu H3378-50 PMTs will be ready at next month.
- Round bottom flask using GE214 is ready. Stored in the clean room
- Inner bag stored for liquid scintillator will be prepared by ETFE sheet. No damage was found.
- We measured the transparency of ETFE sheet using scintillation and obtained 2.5% loss at maximum.
- Centrifuge for ^{96}Zr isotope separation was proposed to JNFL and 25% enrichment maybe possible.

Clean booth and 200g Zr(iPrac)₄

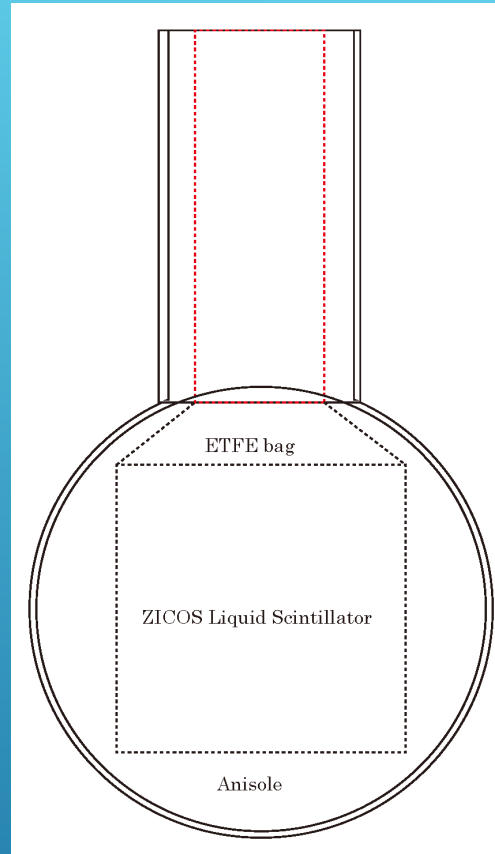


- Glove box with N₂ gas circulation has been installed.
- 200g Zr(iPrac)₄ was stored in dry box.
- 12 Hamamatsu H3378-50 are ready. 8 PMT will be delivered at next month.

Present status

- Clean booth (class 1000) was constructed for preparation of Liquid Scintillator and detector.
- 200g of $\text{Zr}(\text{iPrac})_4$ is ready. Stored in the dry box.
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- Round bottom flask using GE214 is ready. Stored in the clean booth. PMT mounting jig is also designed .
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Ultra-pure quartz flask and inner bag

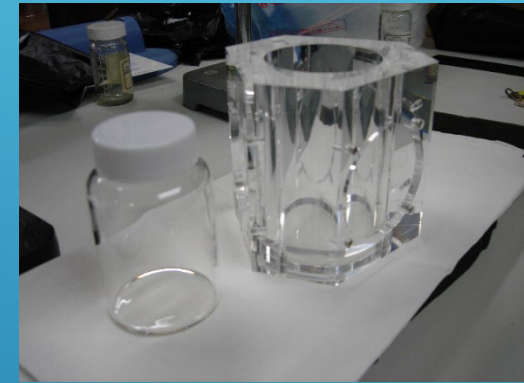
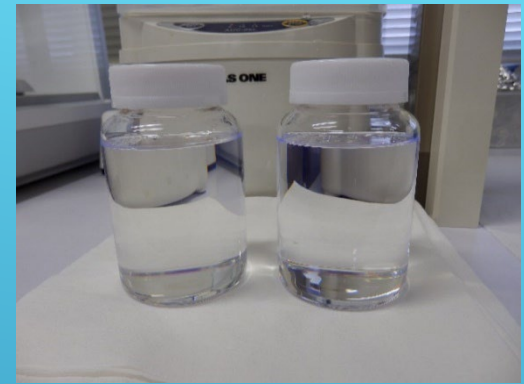
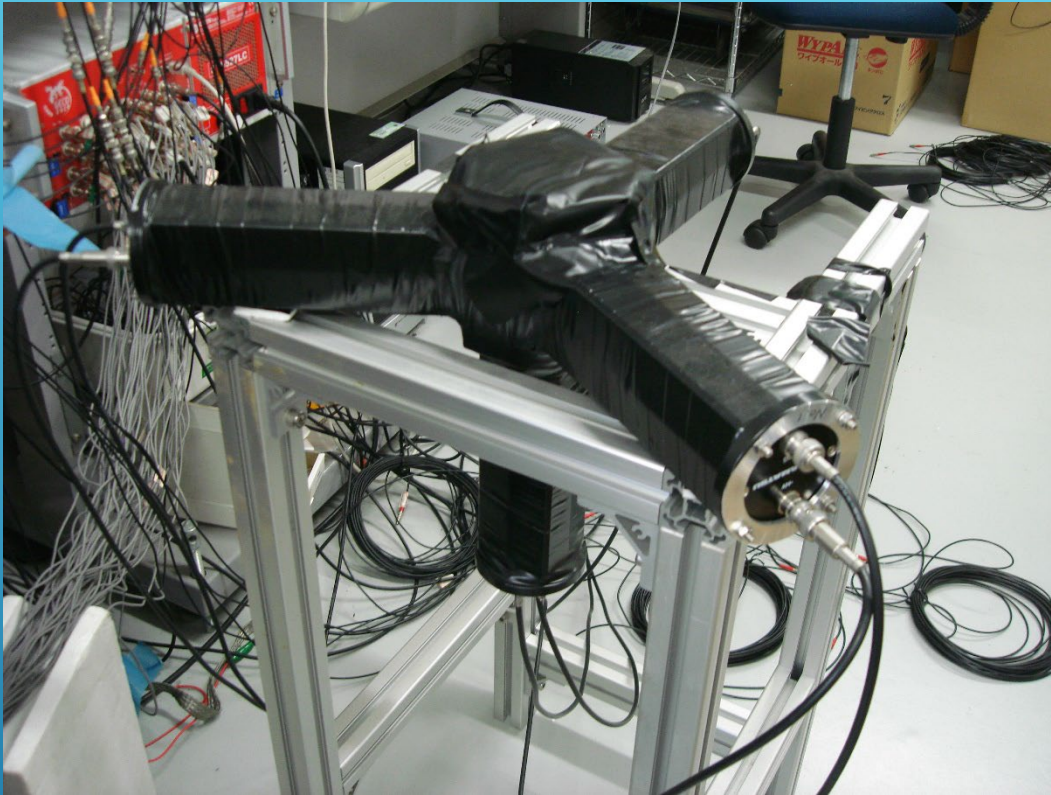


- Ultra-pure quartz round flask is ready.
- We will use ETFE sheet for inner bag. No damage due to Anisole erosion was found within one month.

Present status

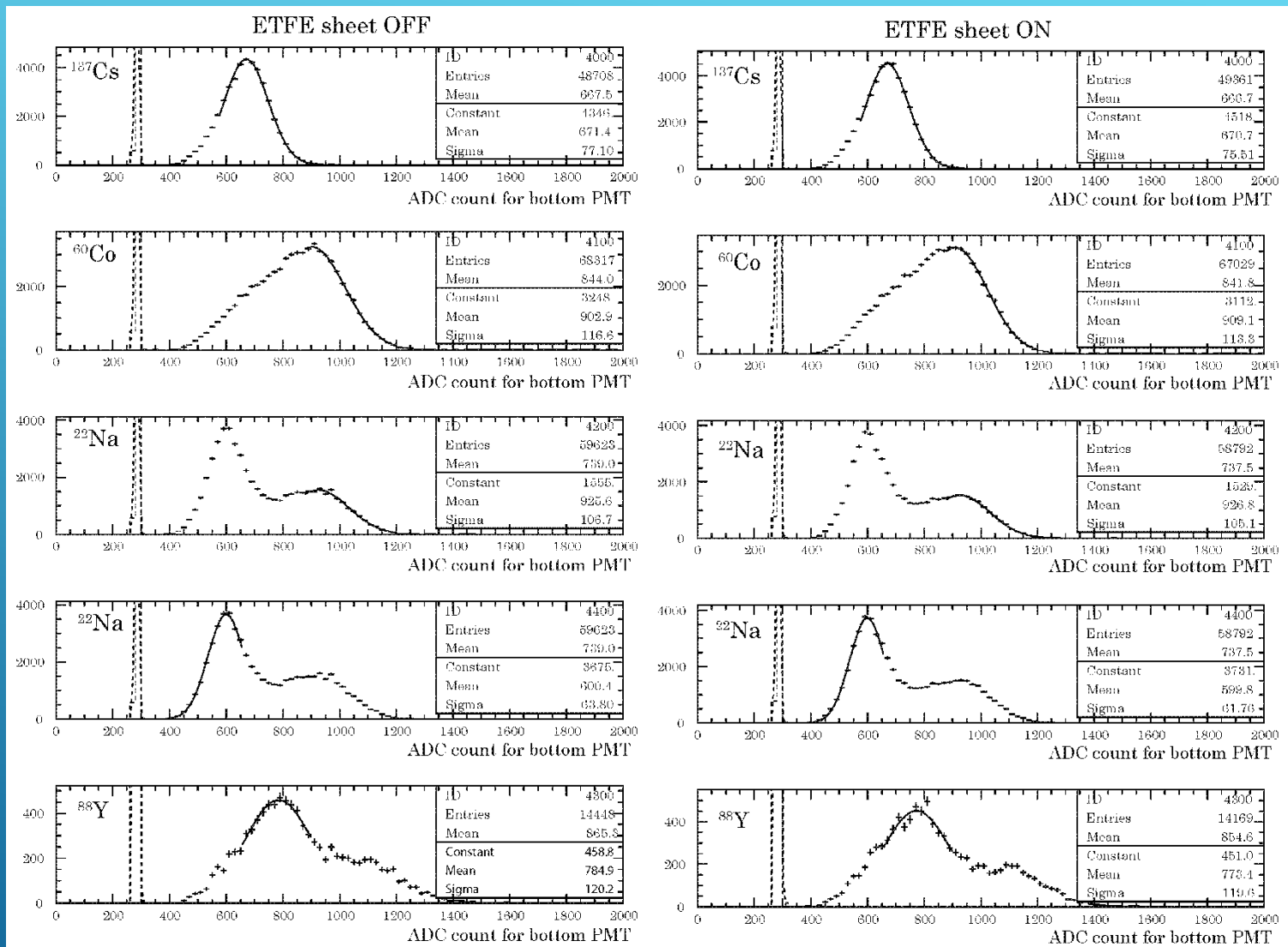
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Measurement of transparency for ETFE sheet



- Transparency of ETFE sheet using Liquid Scintillator.
- Compared peak value of some RIs for side PMTs w/wo ETFE sheet covered side wall of vial.
- Bottom PMT was always without ETFE sheet.

For bottom PMT

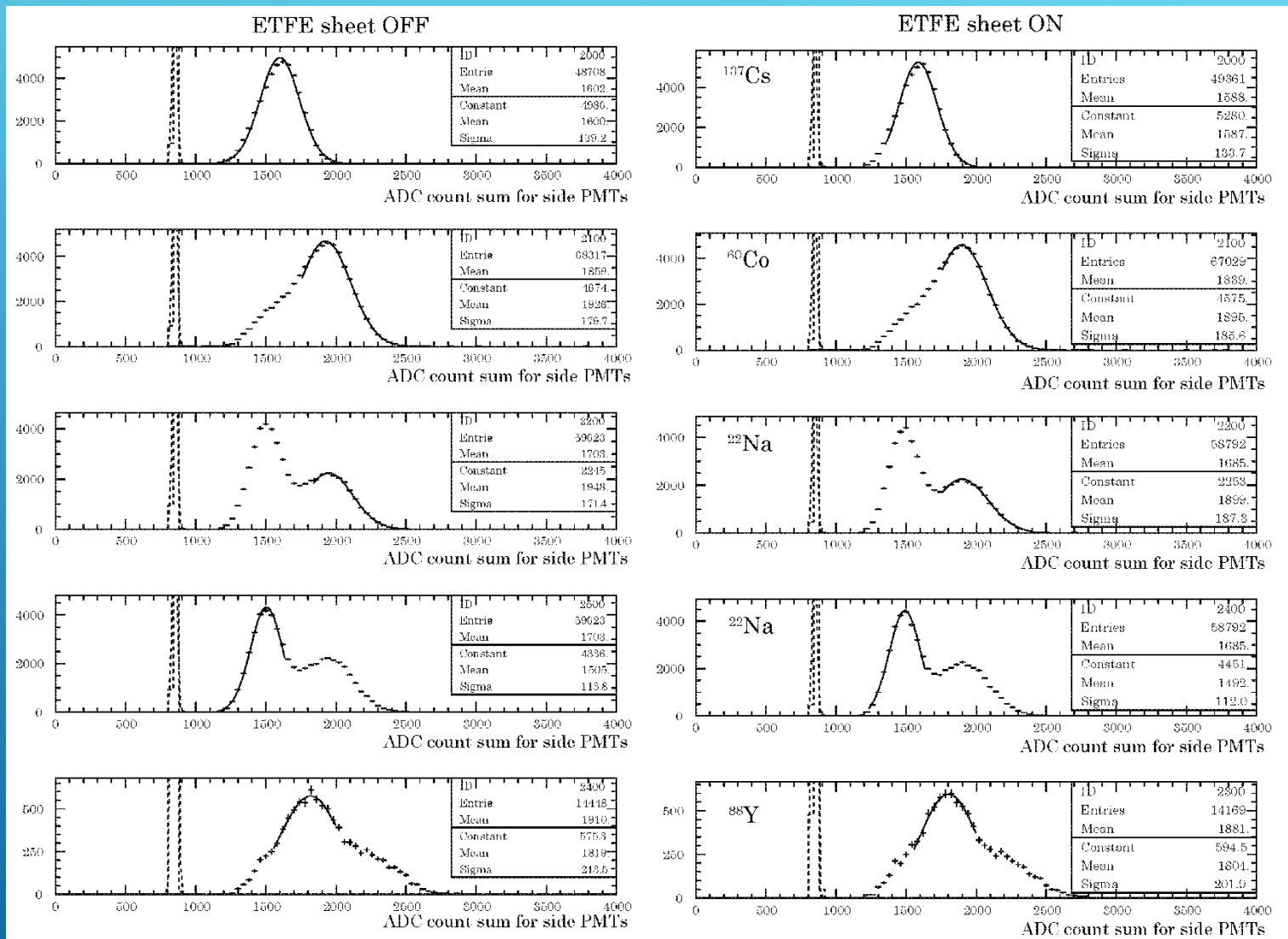


1.0015 ± 0.0018



Same LY as expected

For side PMTs



0.9768 ± 0.0014



2.5% loss at maximum

Present status

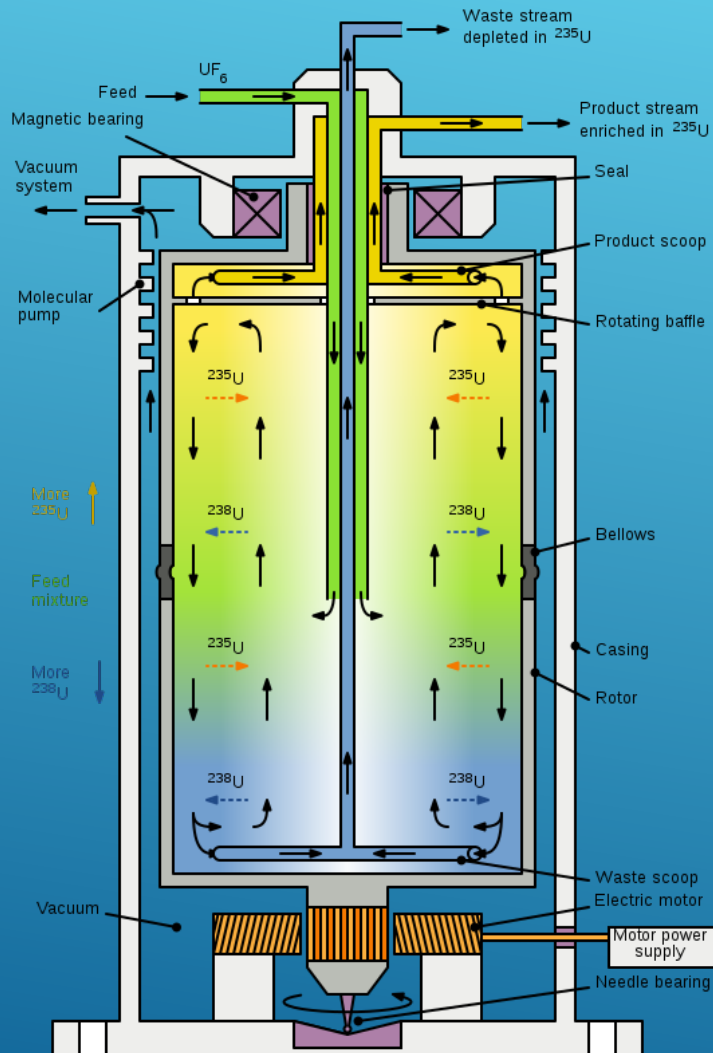
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Centrifuge plant of JNFL at Rokkasho-village



<https://www.jnfl.co.jp/ja/business/about/uran/summary/development-center.html>

^{96}Zr isotope separation by Gas Centrifuge

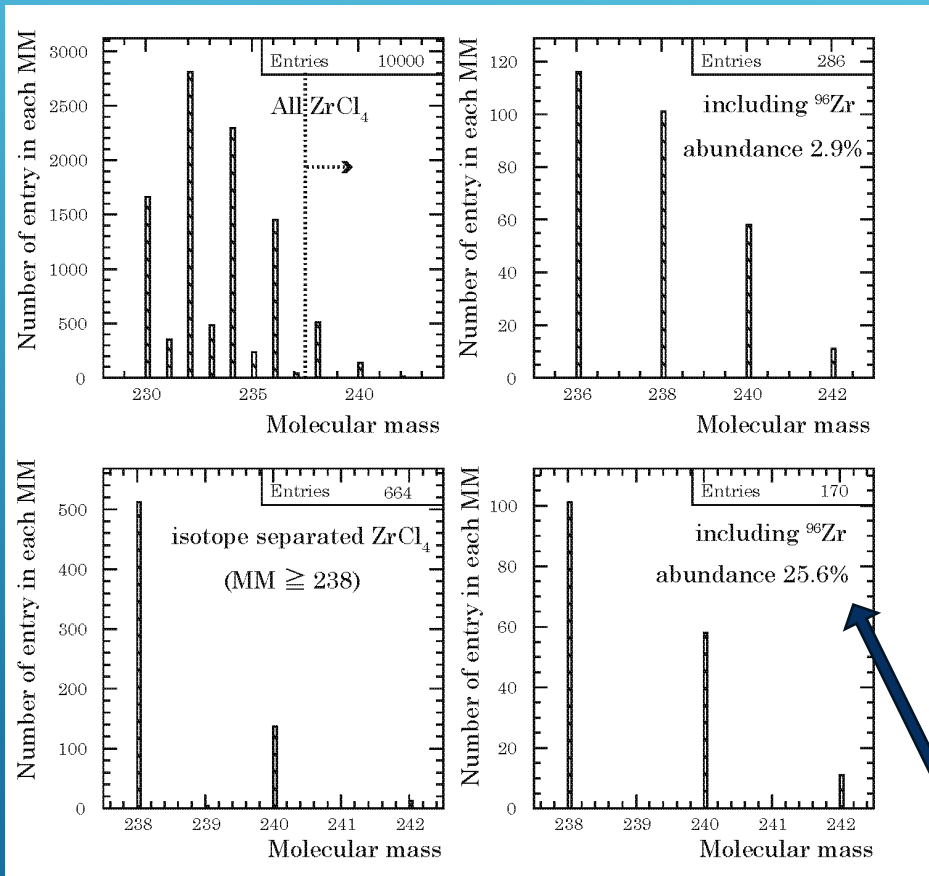


JNFL staffs of Technical Development Center for Uranium Enrichment are looking for new idea for isotope separation using their Centrifuge plant.

Proposed ^{96}Zr separation using ZrCl_4 (bp $331\text{ }^\circ\text{C}$):
Need new sublimation pressure data.

^{96}Zr isotope separation by Gas Centrifuge

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Proposed ^{96}Zr separation using ZrCl_4 (bp 331°C):
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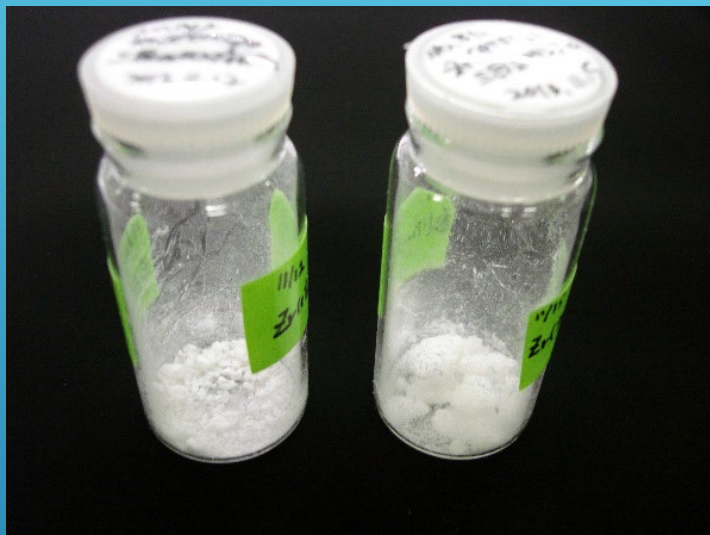
25% enrichment maybe possible by gas centrifuge.

Future plan

- Preparation of ZICOS liquid scintillator will start soon.
- Radiation shielding using Pb blocks with falling prevention wall is designed . Inner size could be 80cm cubic.
- Tuned inner bag using ETFE sheet will be ready after welding test by company.
- Mockup for PMT mounting jig with flask will be done at early next year.
- Installation of inner bag and bringing of LS will be tested by using ultra-pure water after the mockup.
- Construction of Pb shield and 2ν -ZICOS detector will start at next fiscal year in LAB-A.
- **Observation will start in next summer. Stay tuned!**

backup

Zr loaded liquid scintillator



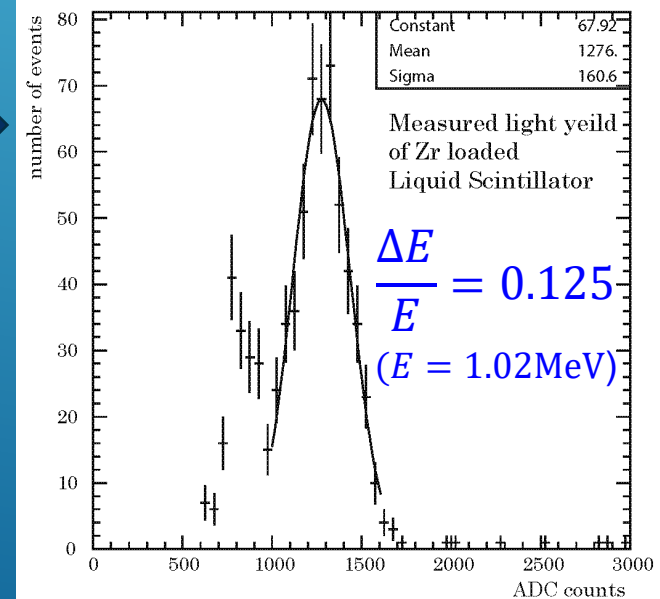
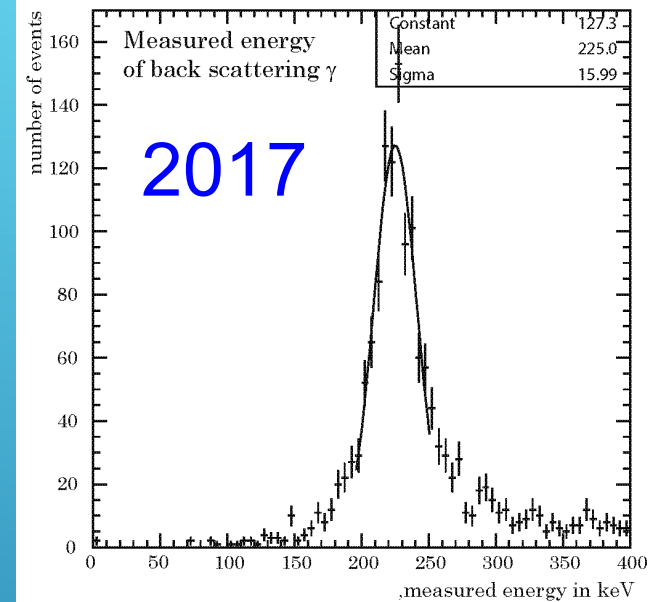
2013~2015

Measured on this August.



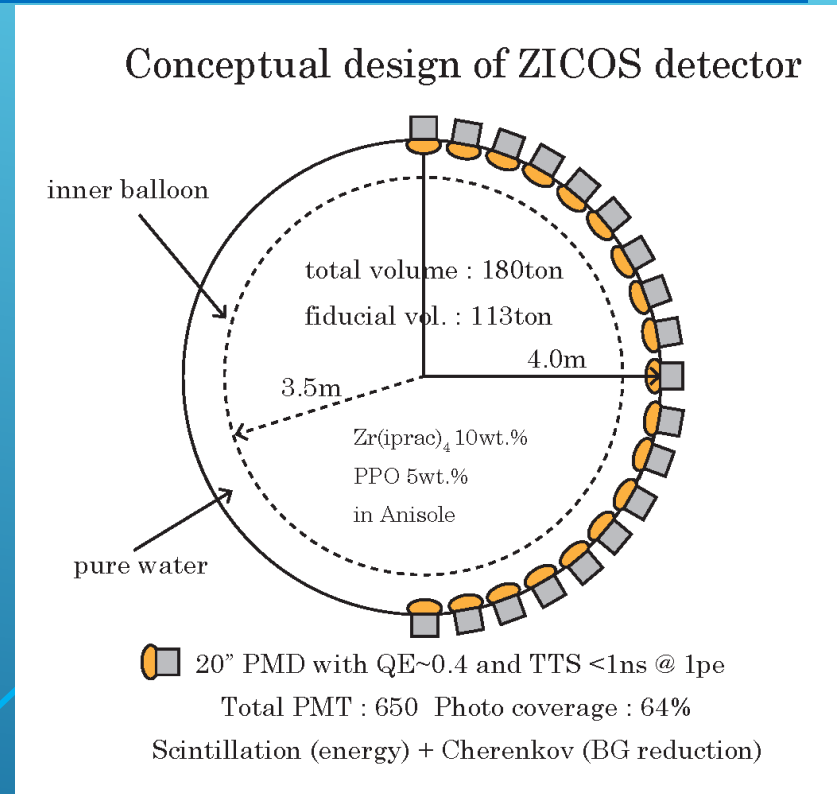
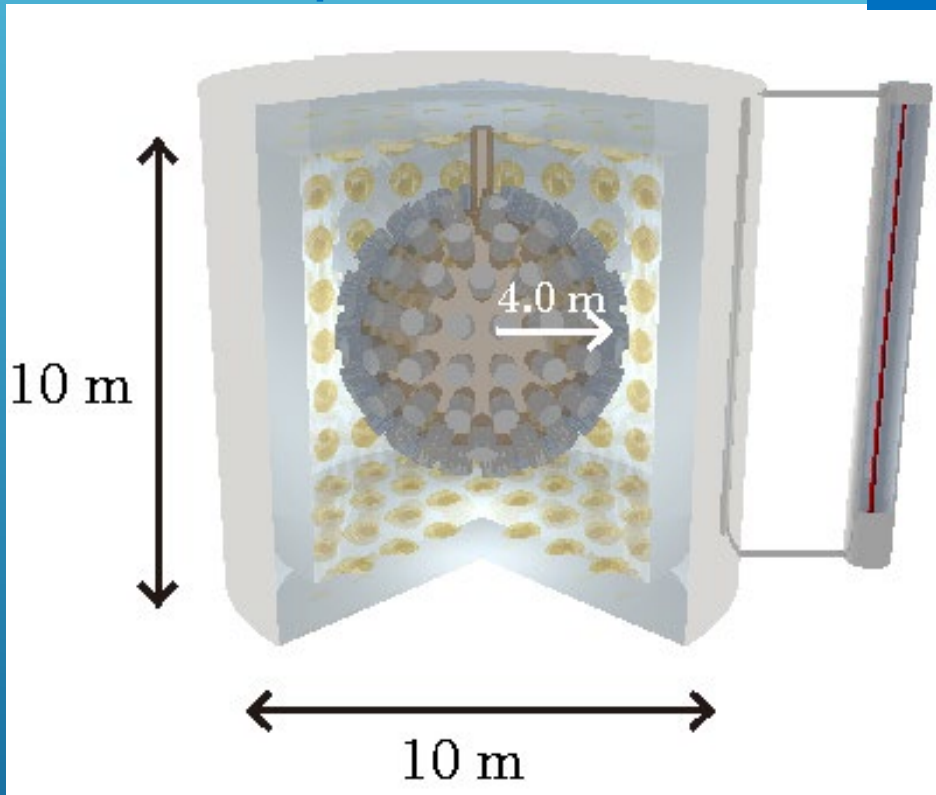
2015

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

NEMO3 : $T_{1/2}^{0\nu} > 9.1 \times 10^{21}$ ys



^{96}Zr : 45kg (nat.) \rightarrow 865kg(50% enrich)
 $T_{1/2}^{0\nu} > 4 \times 10^{25}$ ys \rightarrow $T_{1/2}^{0\nu} > 2 \times 10^{26}$ ys

Experimental site in LAB-A



Need to apply inner-University Research Program

U/Th/K contamination in material

Goal : U/Th ~1ng/g

東芝ナノアナリシス株
材料分析部
JOB No.G800G

【速報】石英ガラス中のU Th量測定

[試料]
石英ガラス
・RQ200
・GE214

計2試料

[方法]
酸分解 – ICP質量分析法 (パーキンエルマー社製 NexION350S)

[結果]
下表に分析結果を示します。

表 分析結果

試料名	K	Th	U
RQ200	330	42	64
GE214	180	15	29
定量下限	5	1	1

【速報】テトラキス (アセト酢酸イソプロピル) シルコニウムと四塩化ジルコニウム中のU/Th量測定

[試料]
・テトラキス (アセト酢酸イソプロピル) シルコニウム
・四塩化ジルコニウム

計2試料

[方法]
試料調製 – ICP質量分析法 (パーキンエルマー社製 NexION350S)

[結果]
下表1,2に分析結果を示します。

表1 分析結果

試料名	K	Th	U
テトラキス (アセト酢酸イソプロピル) ジルコニウム	-	-	-
定量下限	400	50	50

※表中の「-」表記は、定量下限以下であることを示します。

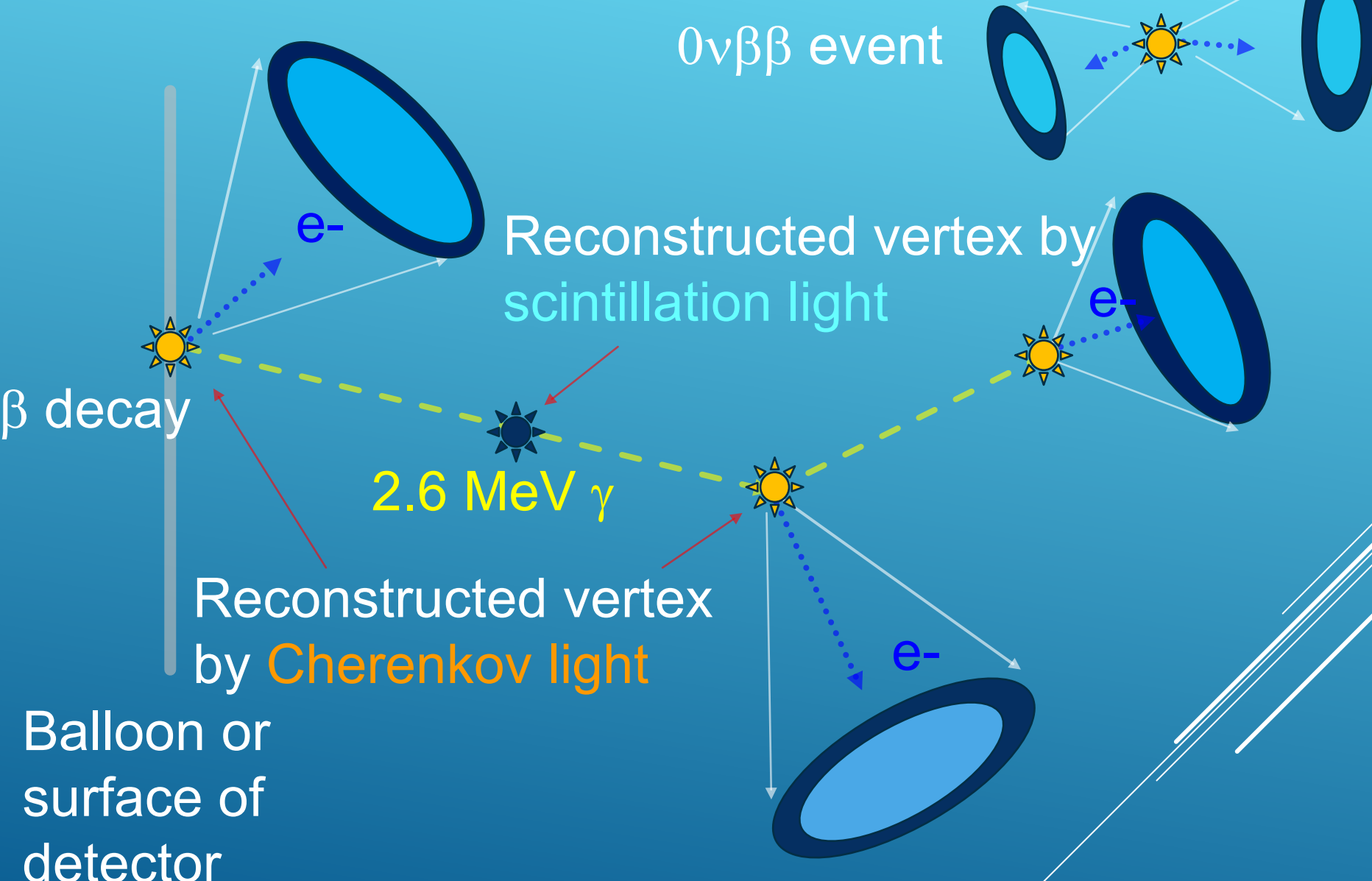
表2 分析結果

試料名	K	Th	U
四塩化ジルコニウム	-	-	-
定量下限	200	50	50

※表中の「-」表記は、定量下限以下であることを示します。

Most serious BGs come from Ultra pure quartz.

Discrimination of signal and BG

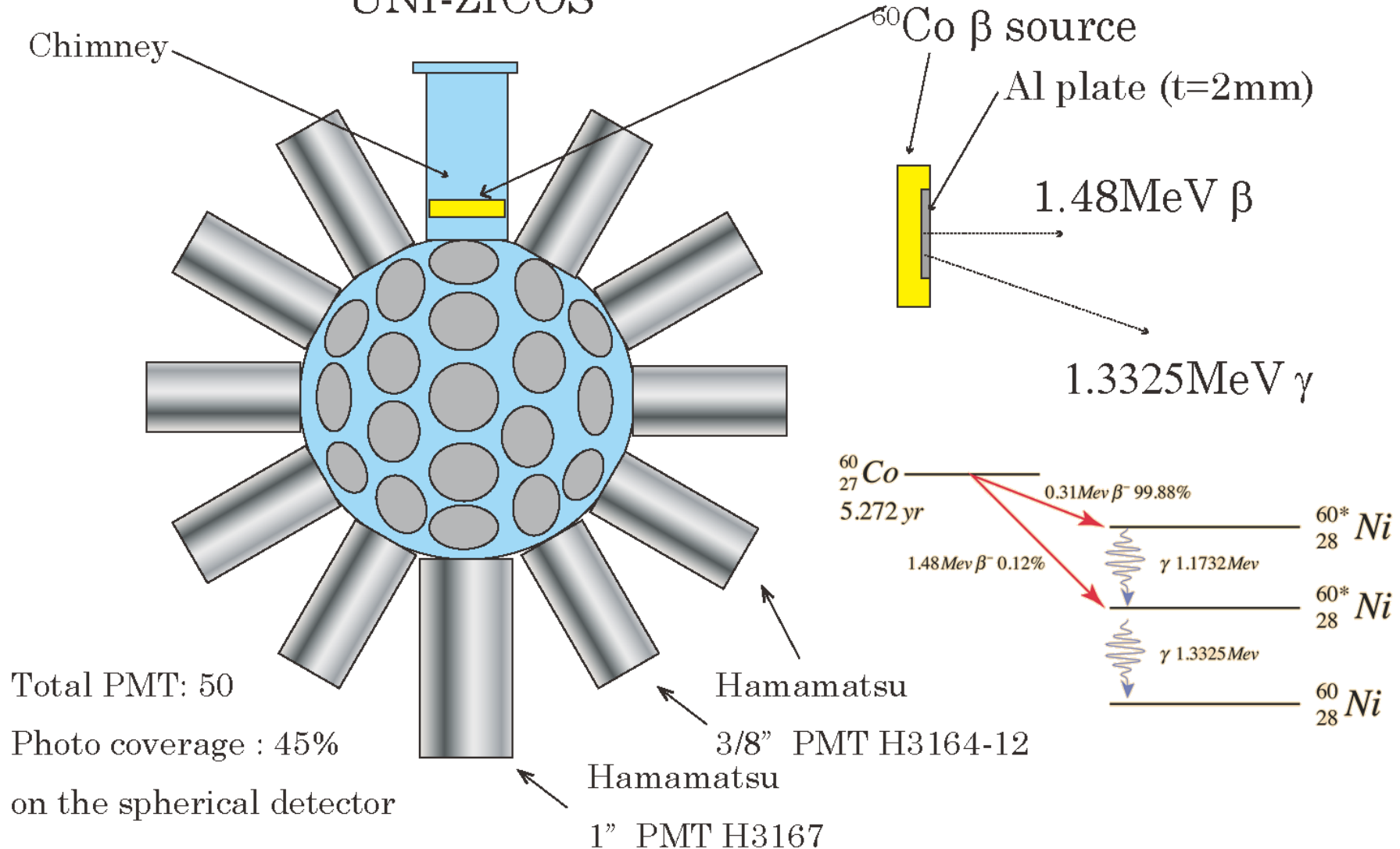


Demonstration of ^{208}Tl background reduction using Cherenkov lights

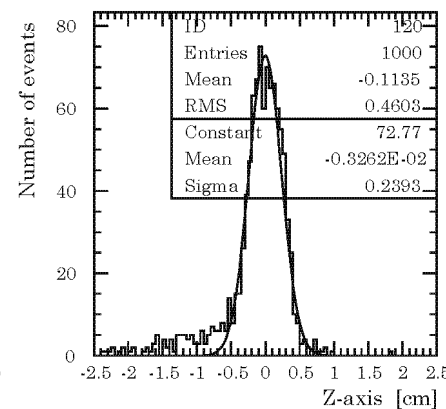
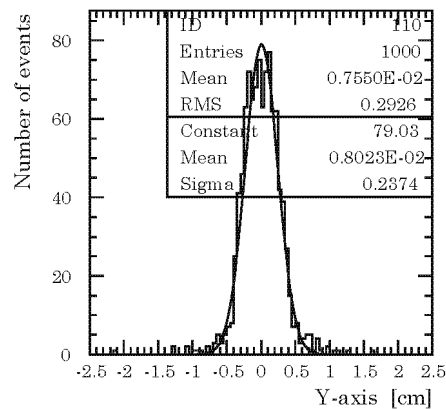
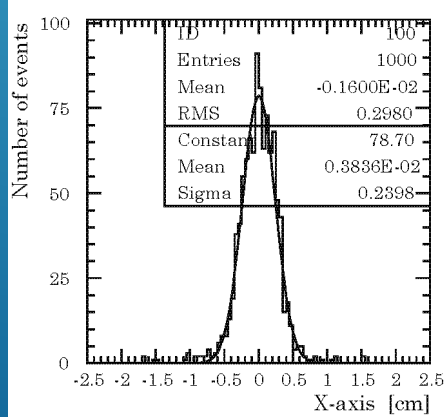
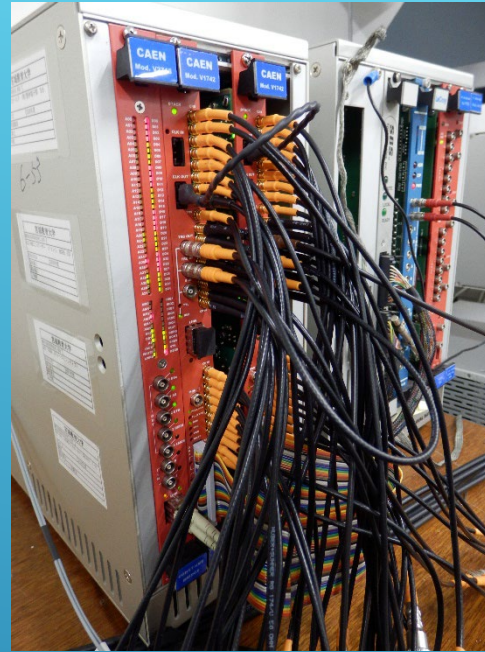
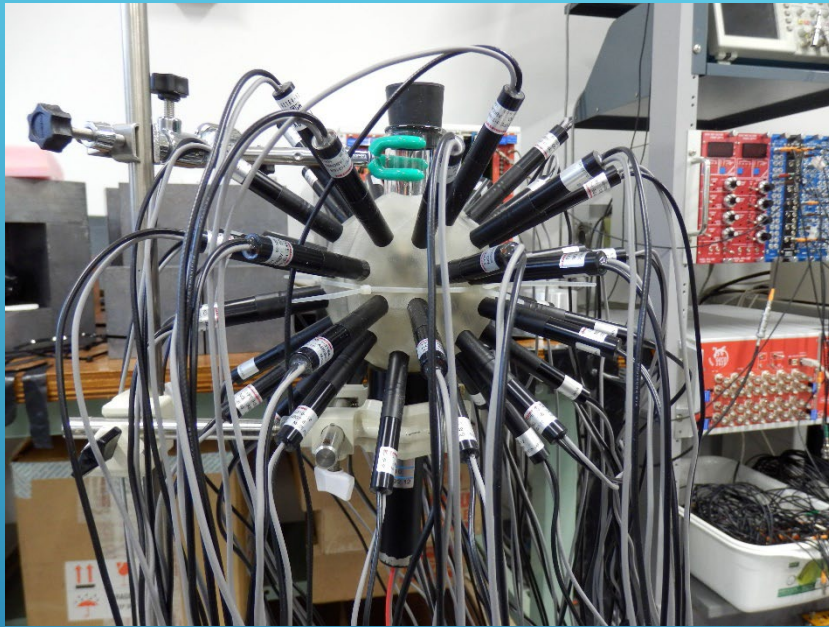
1. Selection of PMTs which receive Cherenkov lights among huge Scintillation lights.
 - Pulse shape discrimination
2. Confirm topology of Cherenkov lights
 - Directionality of Cherenkov lights
 - Direct measurement of topological information as an averaged angle
3. Demonstrate BG reduction using beta-gamma sources with topological information (averaged angle) of Cherenkov light.

Demonstration of ^{208}Tl BG reduction using UNI-ZICOS detector

UNI-ZICOS



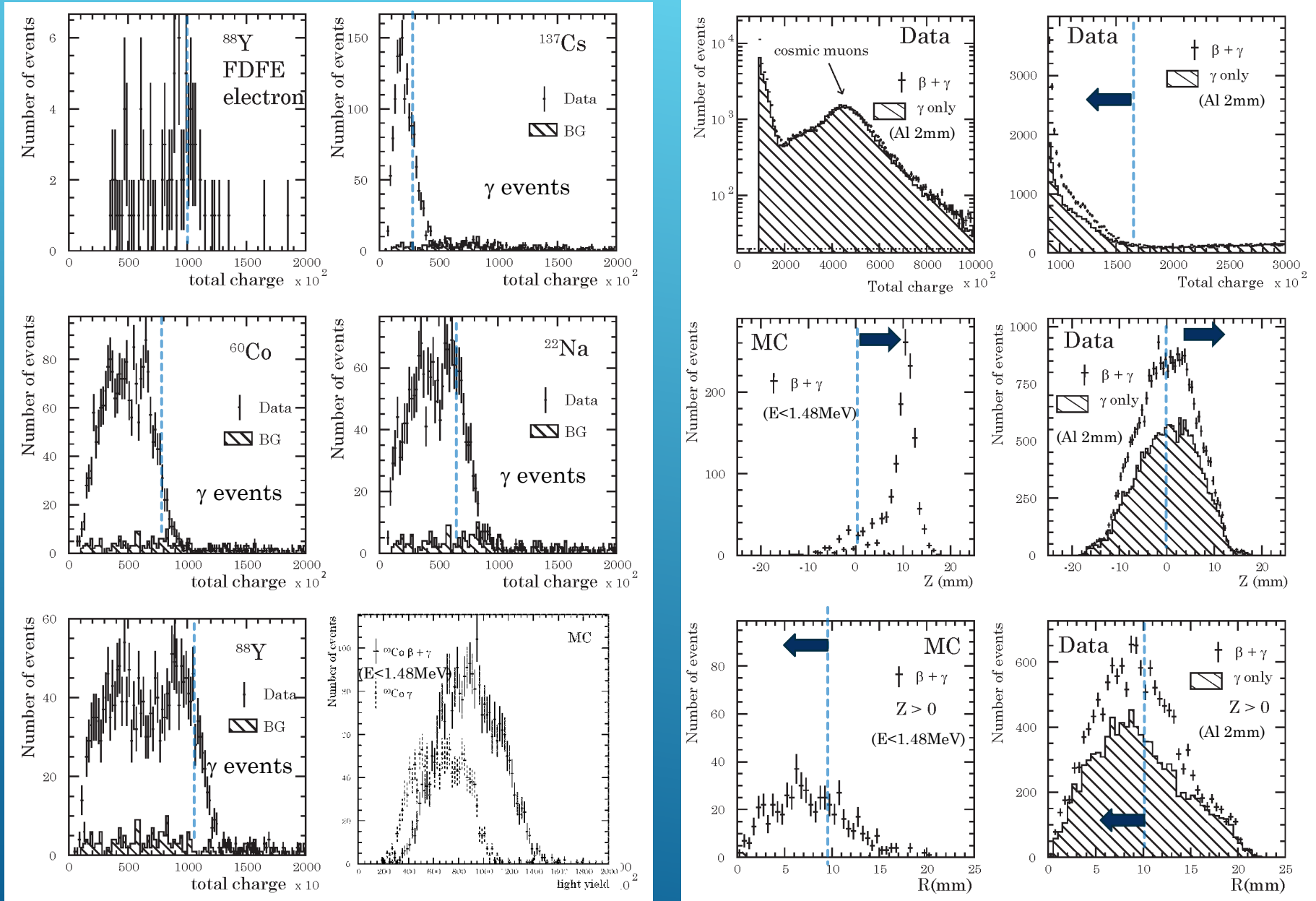
Setup for measurement with UNI-ZICOS



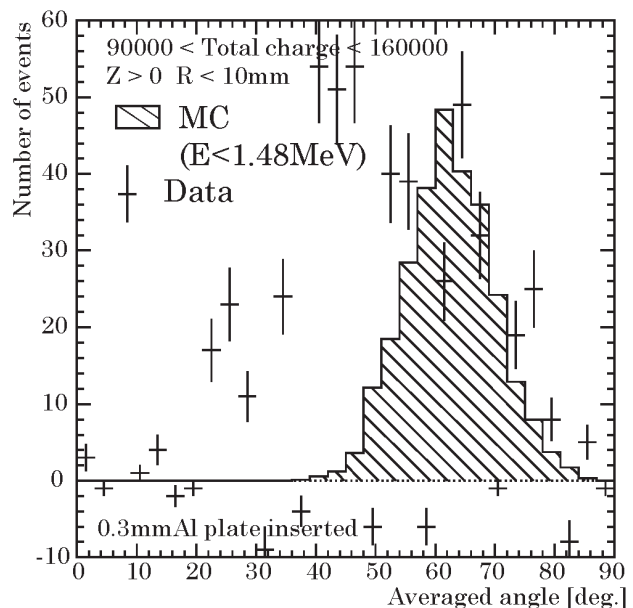
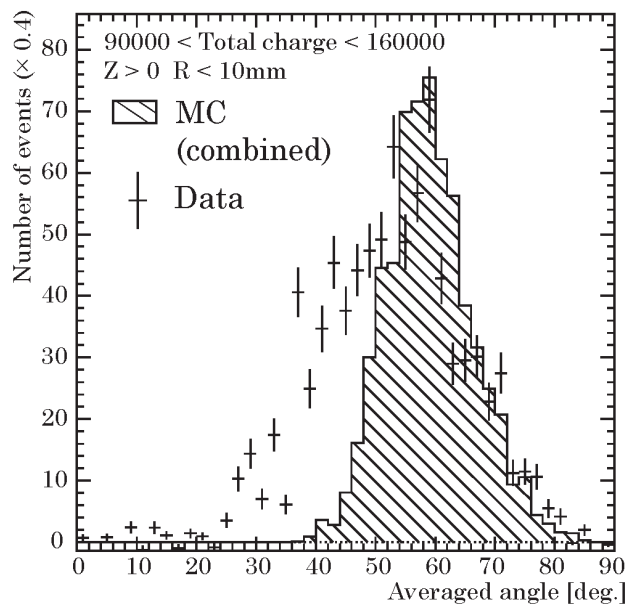
Source holder has a hole, and it can be covered by Al plate (0.3mm and 2mm) to terminate betas.

The vertex position could be reconstructed by assuming that all PMTs should have same effective charge which is corrected by the distance between PMT and vertex.

Events selection for $\beta+\gamma$ rich samples



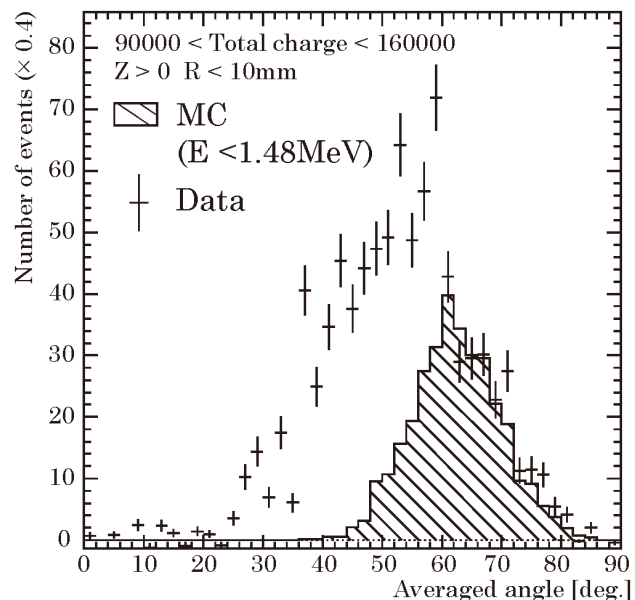
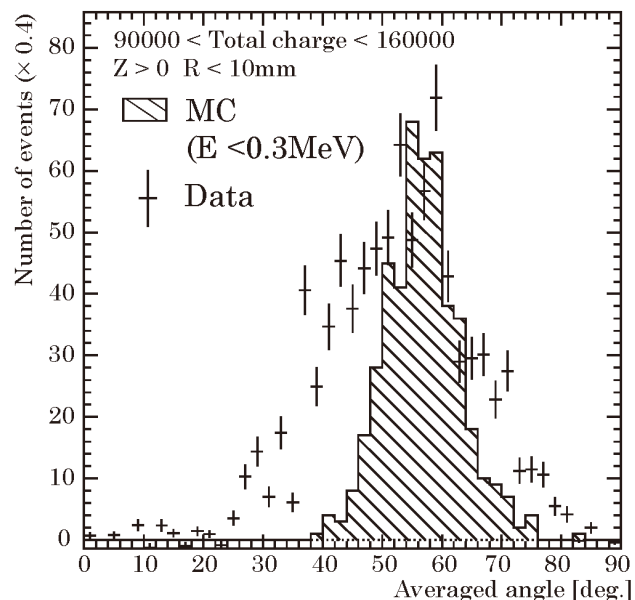
Observed averaged angle using β - γ events



- 0.3mm Al plate terminates β ($E < 0.3$ MeV) events.
- Small difference between 2mm Al on and off
- Obtained averaged angle has small peak around 60 deg.
- There is no clear source to explain around 40 deg events.
- MC($\beta(E < 1.48$ MeV) + γ) looks reproduce higher peak.

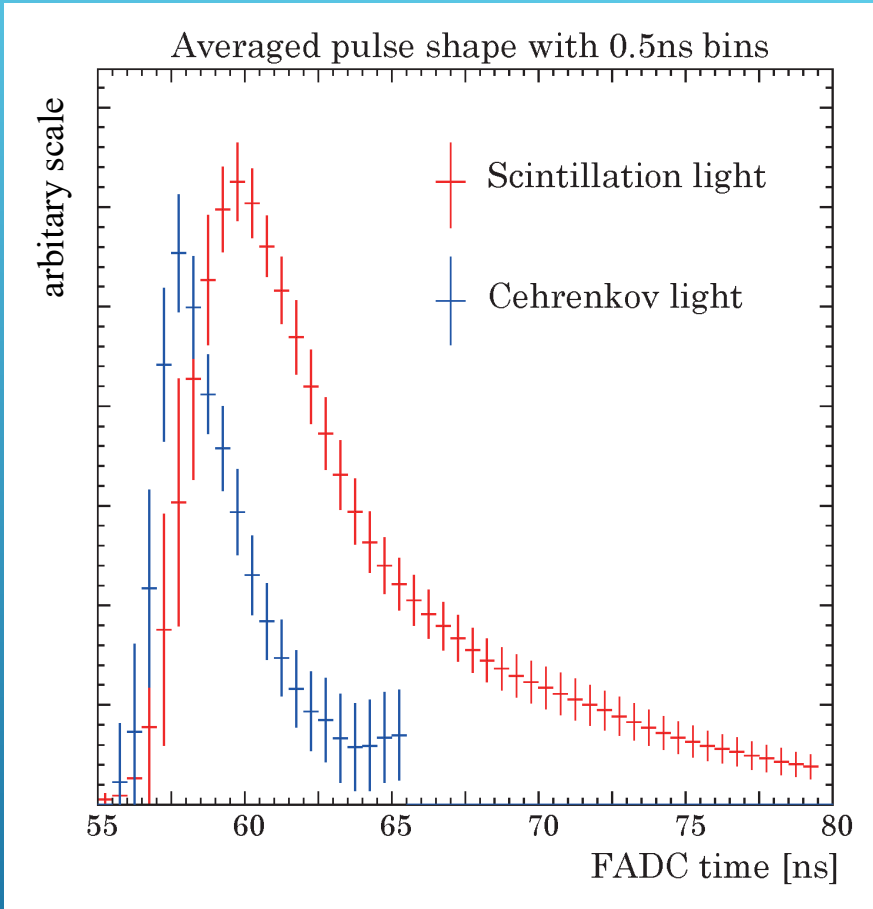
Averaged angle could be used for reduction of $\beta + \gamma$ events such as ^{208}Tl decay background.

Observed averaged angle using β - γ events



- Clear averaged angle of $\beta + \gamma$ events is obtained by subtraction of those data.
- Obtained averaged angle has a peak around 58 deg.
- Averaged angles of MC($\beta(E < 0.3\text{MeV}) + \gamma$) reproduce peak, but not reproduce shape.
- Averaged angle of MC($\beta(E < 1.48\text{MeV}) + \gamma$) may help to reproduce larger angle shape.

Pulse shape of Cherenkov and scintillation

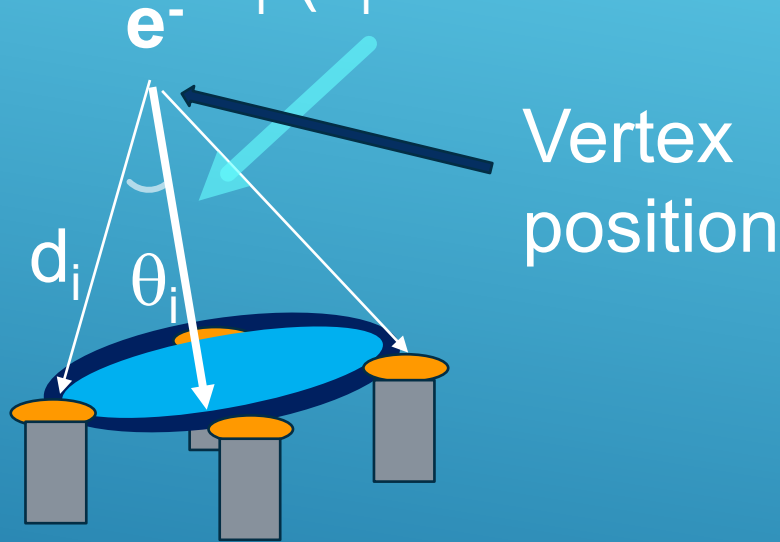


- Pulse shape of ^{90}Sr using H2431-50 measured by V1751 with DES mode (2GS/s)
- Decay time of scintillation : 4.57 ns and 8.38 ns
- Rise time of scintillation : 1.45 ns
- Rise time of Cherenkov : 0.75 ns

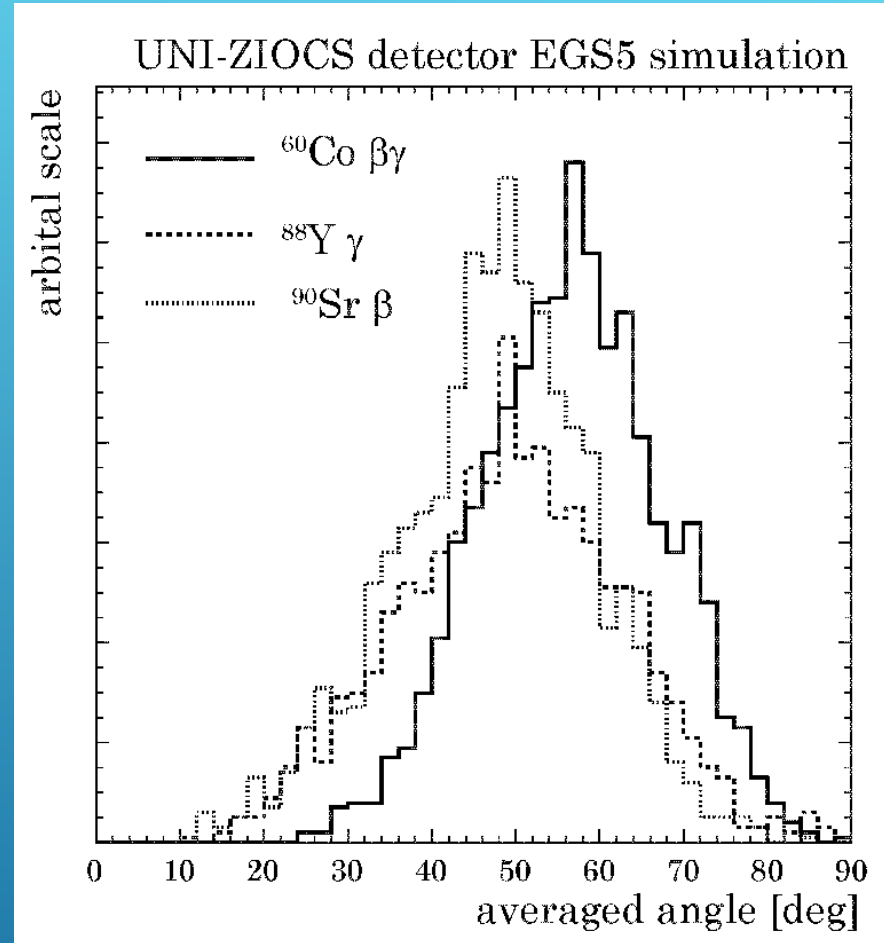
Use the charge ratio $Q_{\text{time}}/Q_{\text{total}}$. Here, Q_{time} is FADC count in each time, and Q_{total} is sum count of FADC between 55 ns and 80 ns.

Topological information of Cherenkov lights

Average direction =
 $\sum d_i$ (d_i : unit vector)

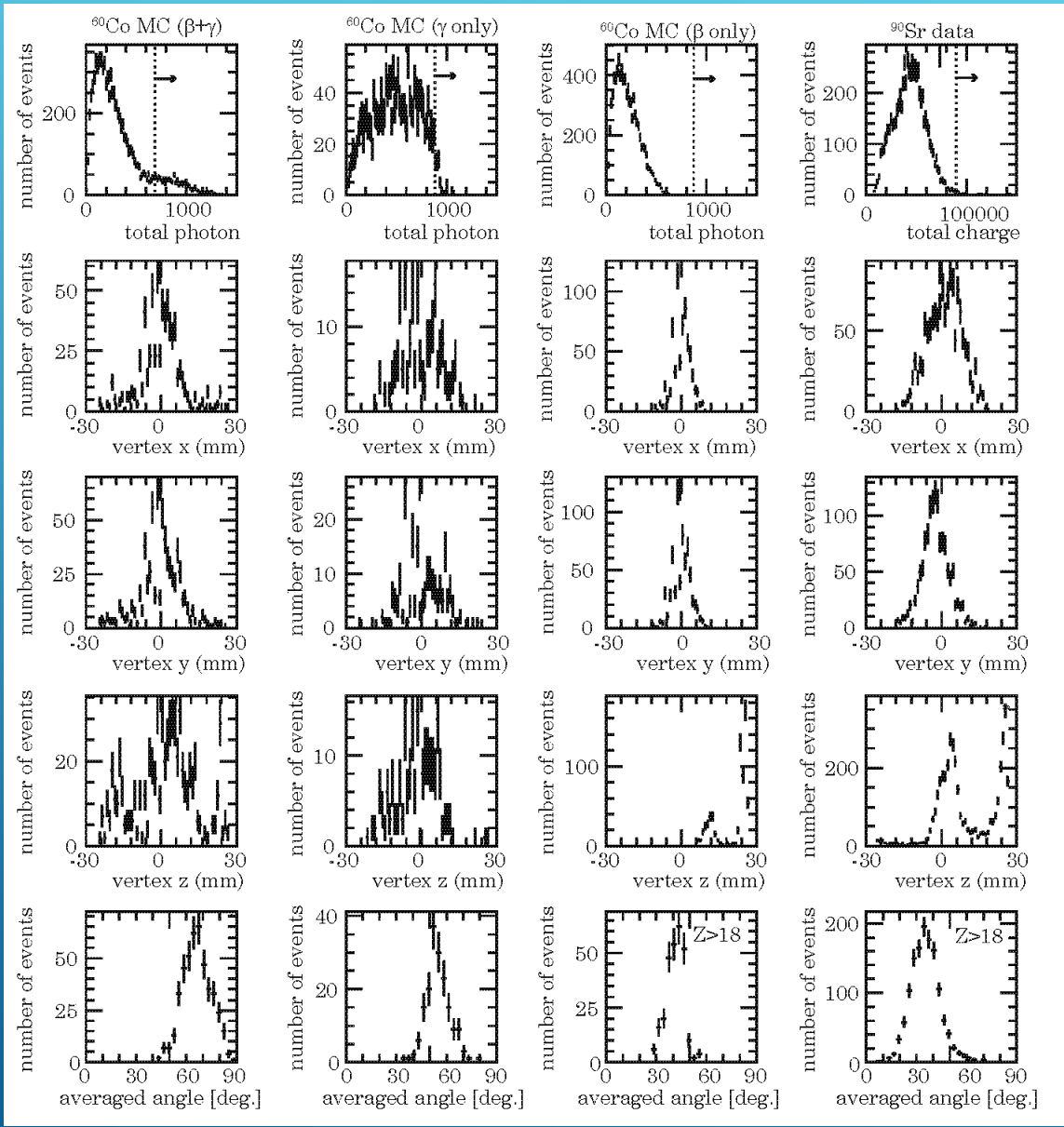


Averaged
angle = $\frac{\sum \theta_i}{N_{hit}}$



Topological information (averaged angle) of Cherenkov lights should be different between β and $\beta + \gamma$ event.

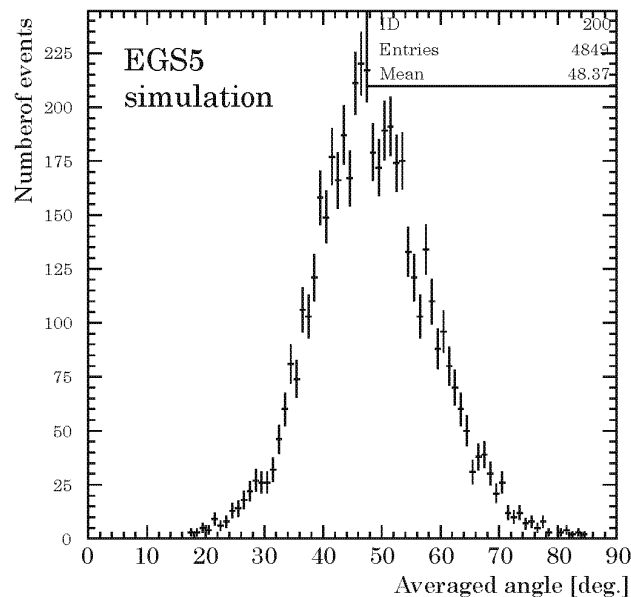
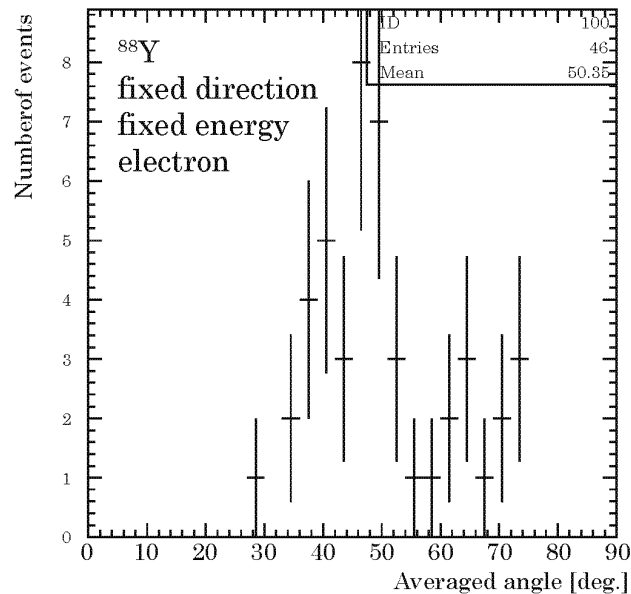
Simulations and data for beta



- Beta MC indicated poor light yield to detect beta only events.
- ^{90}Sr real data confirmed poor light yield even though higher energy than ^{60}Co .

Still there is no clear source to explain around 40deg event.

Observed averaged angle using FDFE events



- The vertex position was reconstructed by charge information as explained.
- Extracted PMTs which include Cherenkov light by PSD technique.
- Using those PMTs and the vertex, the averaged angle of fixed direction fixed energy (1.484MeV) electron from ^{88}Y source was measured, and a peak was found around 50 degree.
- The averaged angle obtained by MC simulation has a peak around 48 degree. Within the statistical error, **the peak position was almost reproduced.**
- Measurement time was limited by March earthquake in Japan.



ZrCl₄分散後



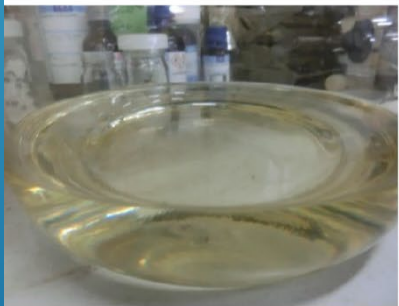
アセト酢酸イソプロピル



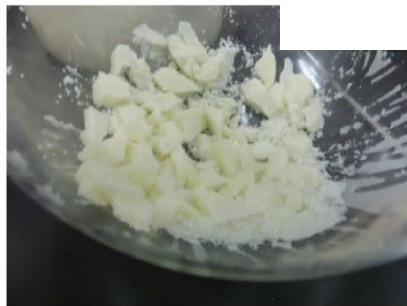
トリエチルアミン滴下後



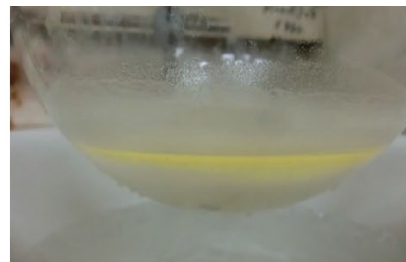
反応終了時



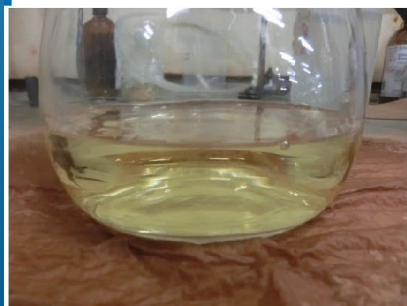
ろ過後ろ液



ろ液濃縮後



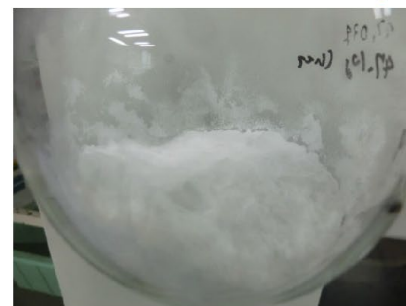
再結晶の様子



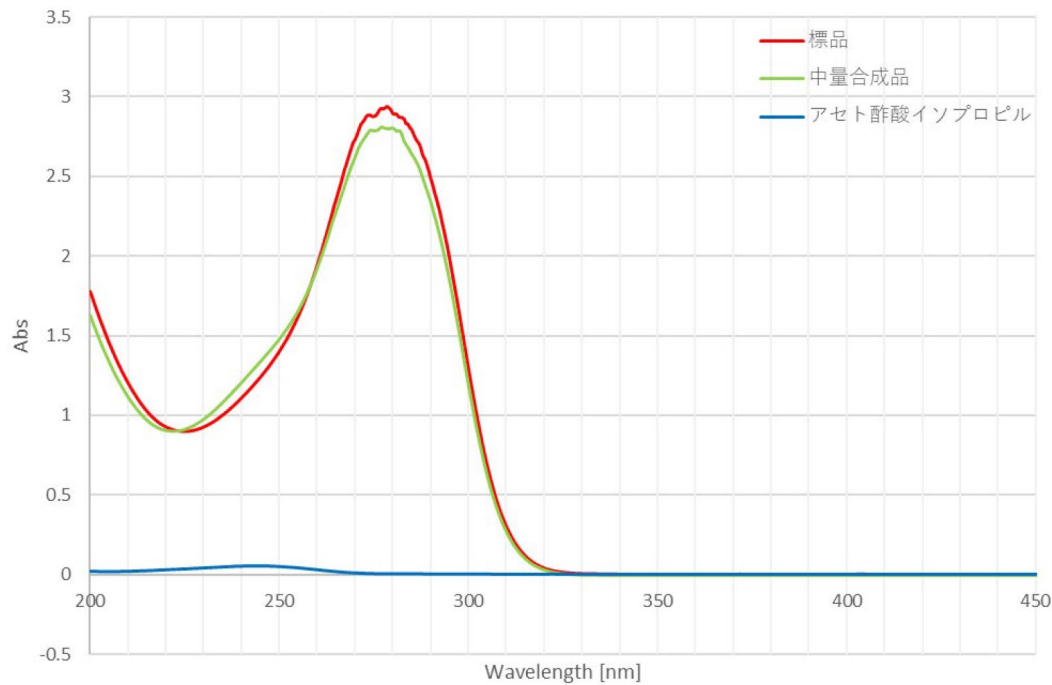
再結晶ろ液



再結晶ろ物



ろ物乾燥後

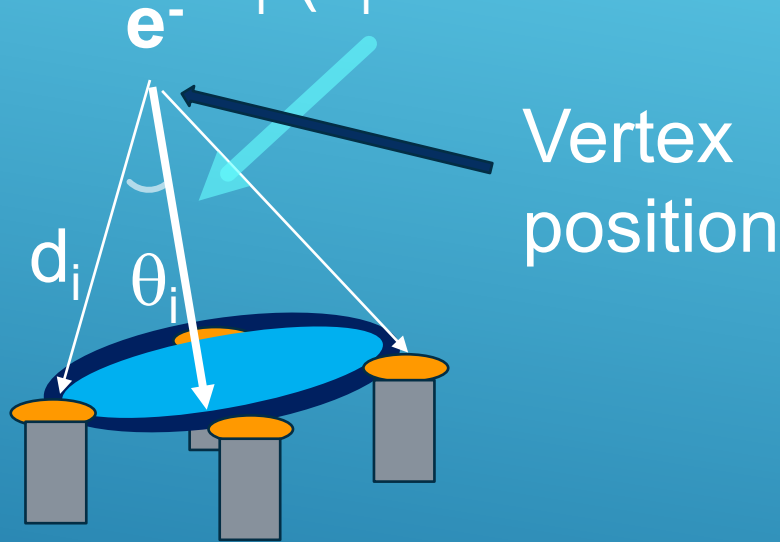


Synthesis of
Zr(iPrac)
was
succeeded.

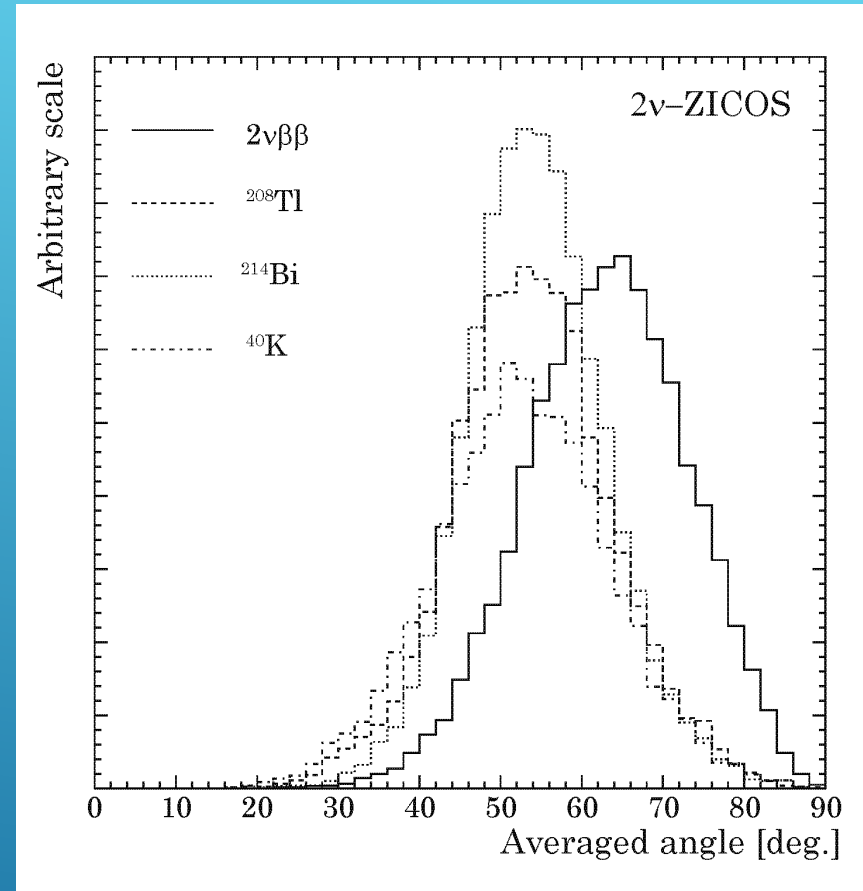
August 29, 2023

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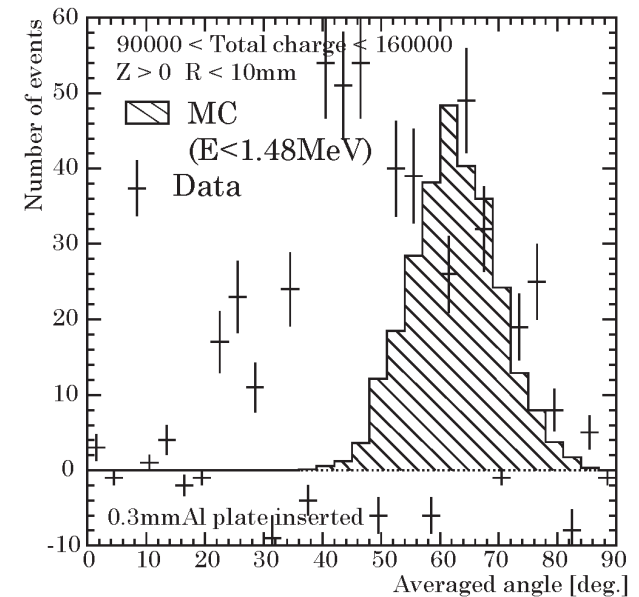
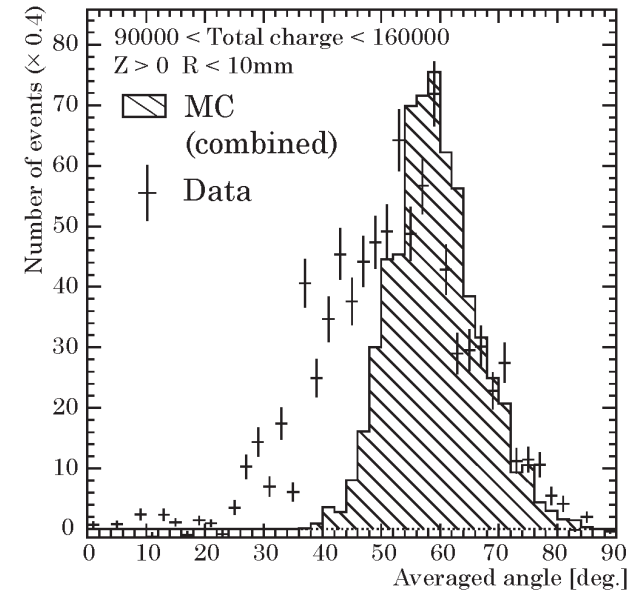
Topological information (averaged angle) of Cherenkov lights should be different between $2\nu\beta\beta$ and $\beta + \gamma$ event.

Topological information of Cherenkov lights



MC($\beta + \gamma$) does not reproduce the small bump around 40 degree.

They might be caused by beta only events which don't have enough light yield as indicated by MC. (at last JPS)



Results of test synthesis of Zr(iPrac)₄ by NARD

¹H-NMR測定結果

NARD

[¹H-NMR, 400 MHz, CDCl₃]

アセト酢酸イソプロピル

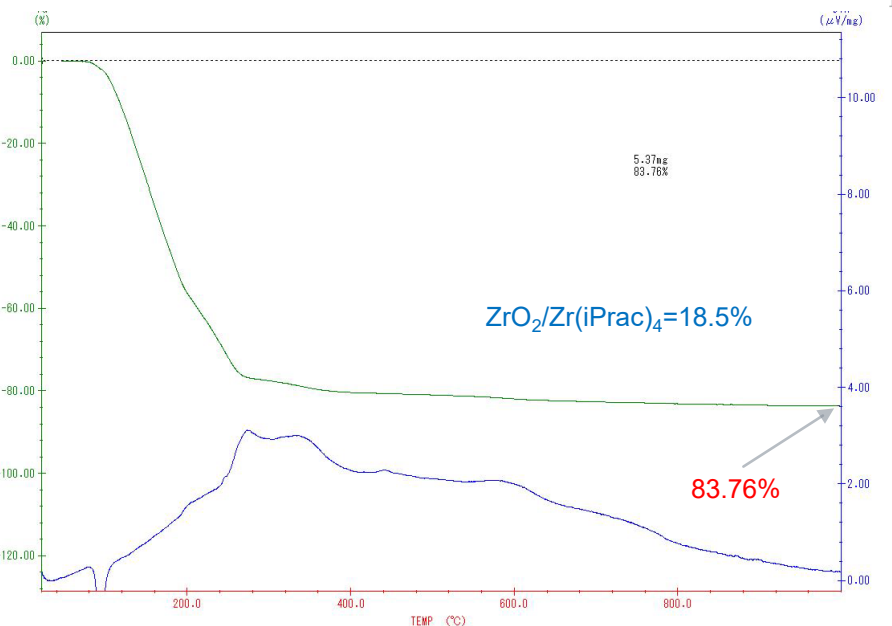
トレース実験 crude

再結晶後 (Hexane)
(heat 40 °C → cool (-20 °C))

再結晶後 (IPA)
(heat 40 °C → cool (-20 °C))

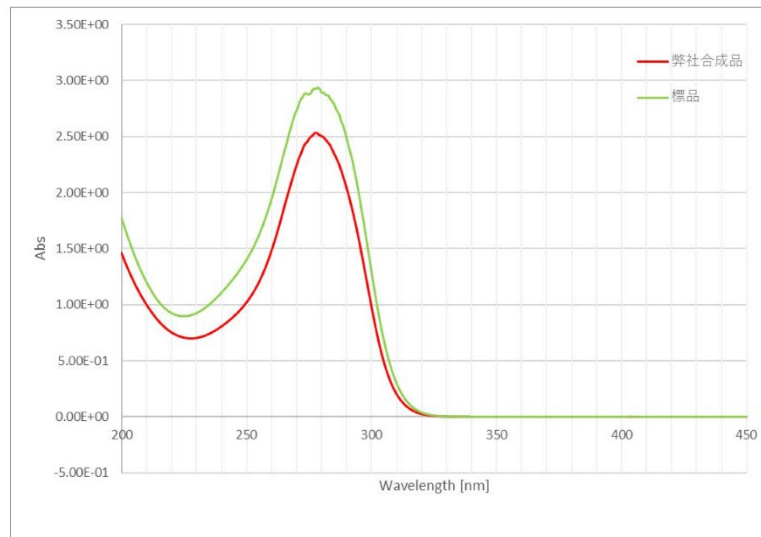
全量再結晶 (Hexane)
(heat 40 °C → cool (r.t.))

PPM



UV測定結果 [送付いただいた標品との比較]

NARD
Confidential



UV測定の結果、弊社合成品の極大吸収波長は278nmであり、標品と同様の結果であった。

Synthesis of Zr(iPrac)₄
was succeeded.