

Development of low- background photomultiplier tubes for future liquid xenon detectors

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Nagoya Workshop on Technology and Instrumentation in
Future Liquid Noble Gas Detectors

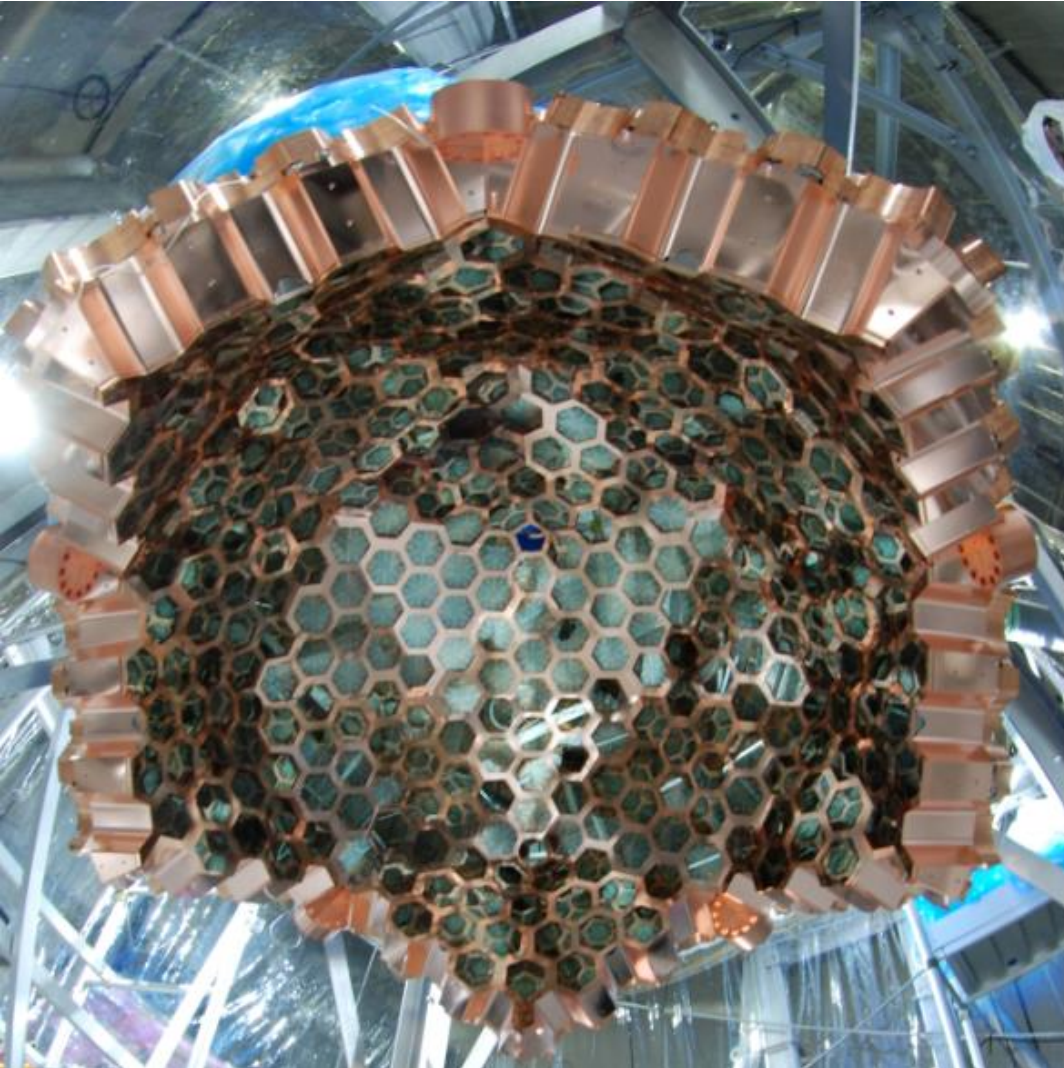
K. Abe

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K. Abe et al. (XMASS Collaboration), JINST 15(2020) P09027, [arXiv:2006.00922 \[physics.ins-det\]](https://arxiv.org/abs/2006.00922)



Introduction



- PMT, one of the candidate as a photo sensor for the future low BG experiment
 - Used in many experiments.
 - Old but stable and well understood technique.
 - In many cases, it is one of the largest RI source.
 - Large volume and material amount.
 - Some parts are difficult to change to keep performance.
 - Lowering RI is very important issue.
- Ultra low background PMT, R13111
 - For LXe detector
 - Low RI

Development of R13111

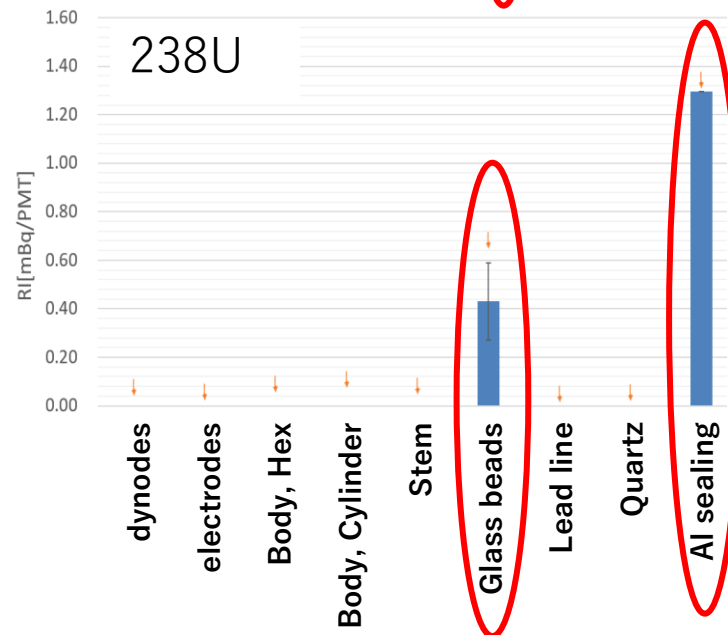
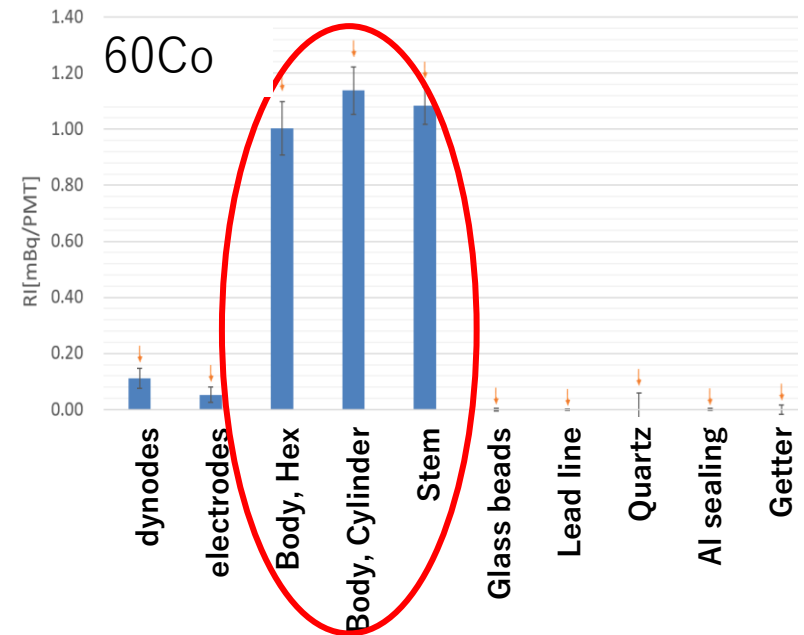
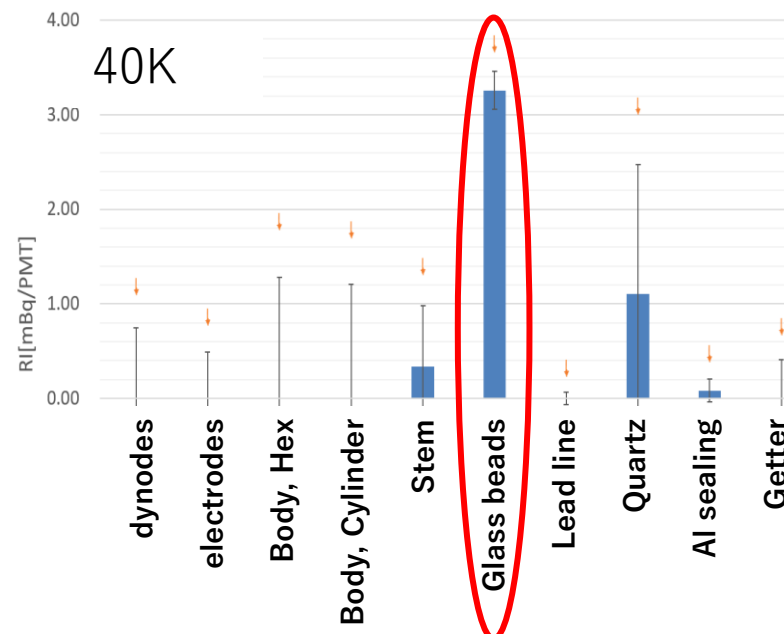
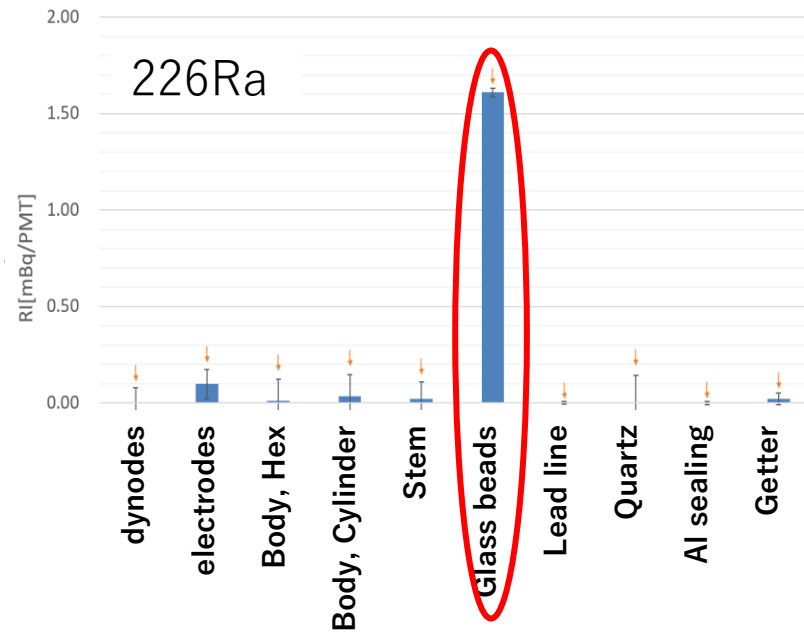
- Developed for future liquid Xe detector based on R10789 with Hamamatsu
 - R10789
 - 2 inch
 - Hex shape photo-cathode to cover the area efficiently.
 - Low RI PMT developed for XMASS-I.
 - ~mBq/PMT amount of radioactivity for each RI
 - Still one of the largest BG source in XMASS-I.
- 3 inch diameter, concave photo-cathode
 - For effective surface event identification.
 - Good TTS
 - $2.11+0.31-0.18$ ns
 - large improvement from R10789 $6.87+/- 0.31$ ns
 - Photo cathode shape and optimized dynode structure.
- RI reduction
 - in following page



RI reduction

- Done based on R10789
 - Listed items which has large contribution in R10789
 - Main target to reduce RI
- All components of PMT were screened and selected by RI measurements.

RI in each component of R10789




- ^{226}Ra , ^{60}Co , ^{40}K , ^{238}U
- Large contributions are from
 - Glass beads
 - Insulators for feedthrough
 - $^{226}\text{Ra}/^{228}\text{Ra}/^{40}\text{K}/^{238}\text{U}$
 - Kovar metal parts for body and stem
 - ^{60}Co
 - Kovar metal has good property to connect to quartz window, but it includes cobalt.
 - Al sealing is another RI source.
 - ^{238}U

Large contributors in R10789

- From each component measurement
 - Glass beads
 - Kovar metal
 - Aluminum seal
- From assembled PMT measurement
 - photocathode production was identified as largest 40K source.
 - $> \sim 6\text{mBq/PMT}$ and largest contribution for 40K.
 - cannot be fixed by each component measurement.

Samples	^{226}Ra	^{228}Ra	^{40}K	^{60}Co
Assembled PMT (R10789)	1.2 ± 0.3	< 0.78	9.1 ± 2.2	2.8 ± 0.2
Sum of the PMT parts (R10789)	1.6 ± 0.3	1.1 ± 0.3	< 3.2	3.4 ± 0.2

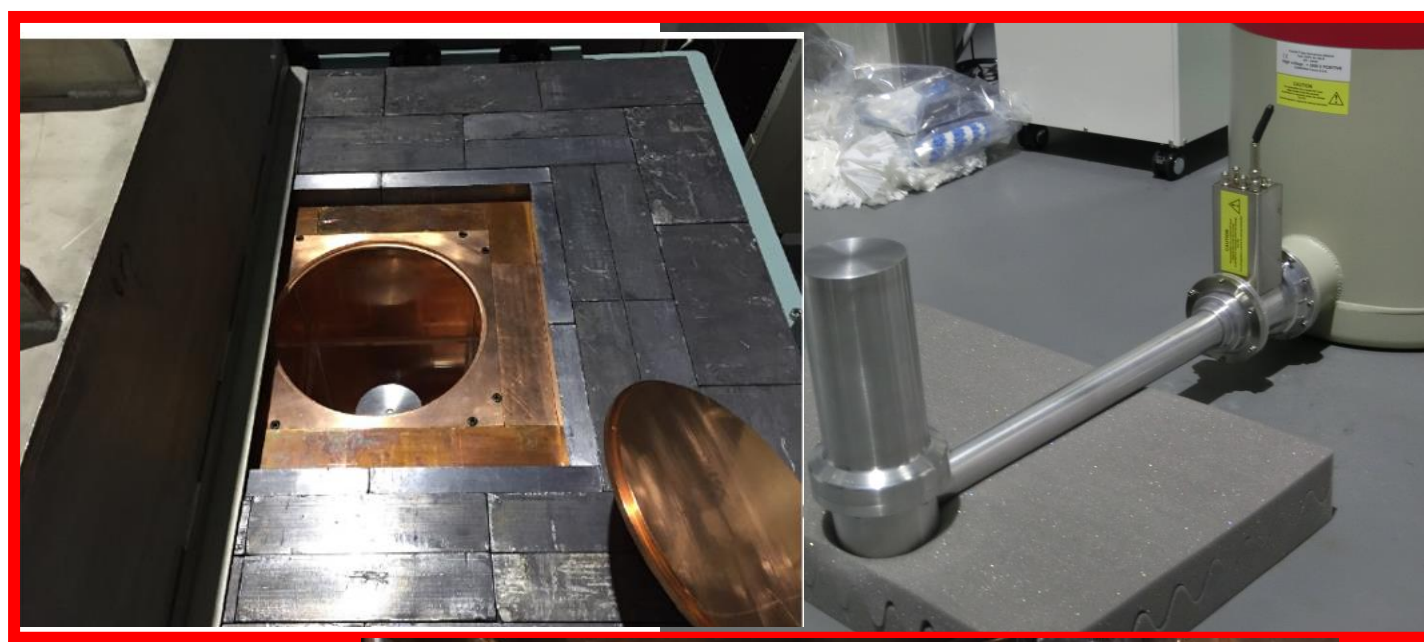
contamination during assemble 

RI screening

- All components of PMT were screened and selected by RI measurements.
- RI measurement were done by HPGe and mass spectrometer
- HPGe
 - 0.01 mBq/PMT
 - Increased sensitivity from R10789 development
 - Increase amount of samples in one measurement.
 - New low BG Ge.
 - systematic +30% -10%
 - calibration data and simulation difference
- Mass spectrometer
 - HPGe has less sensitivity for some RI such as upper stream of decay chain, ^{238}U and ^{232}Th , ^{40}K .
 - For those RI, mass spectrometer often has better sensitivity
 - If the volume of required material for one PMT becomes large
 - 2 types of mass spectrometer was used
 - ICPMS, systematic +/- 10%
 - GDMS, systematic +/- 30%

HPGe detectors

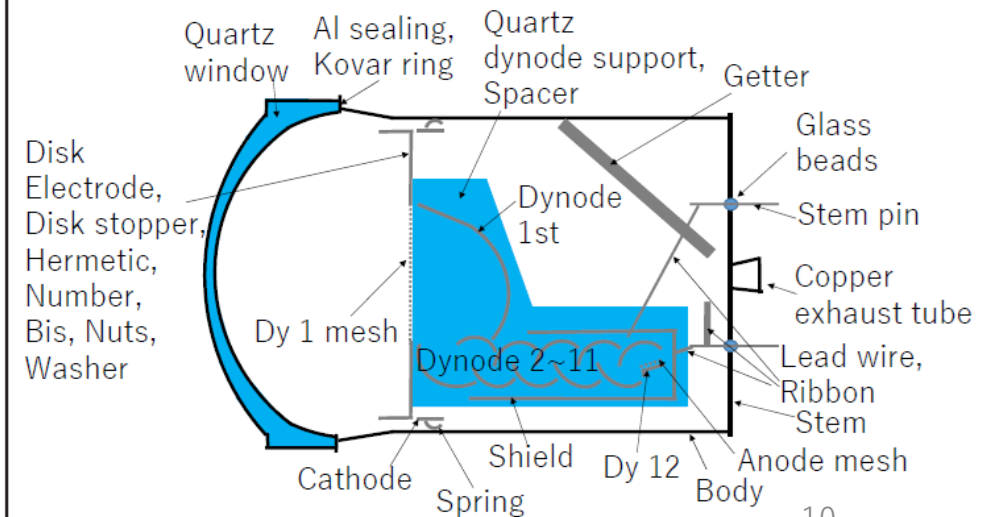
- Ge detectors used for this screening.
 - 3 p-type detectors
 - 1.9 kg, 100%, relative efficiency
 - 2.5 kg, 120%, diameter 83mm, height 86mm
 - 1.7 kg, 80%
 - Specially prepared for low RI measurement.
 - Large sample chamber to measure large sample and large amount, such as assembled PMT.
 - n-type
 - 1.9 kg, 100%, diameter 80.5mm height 80.5mm



Components, amount of measured samples for R13111

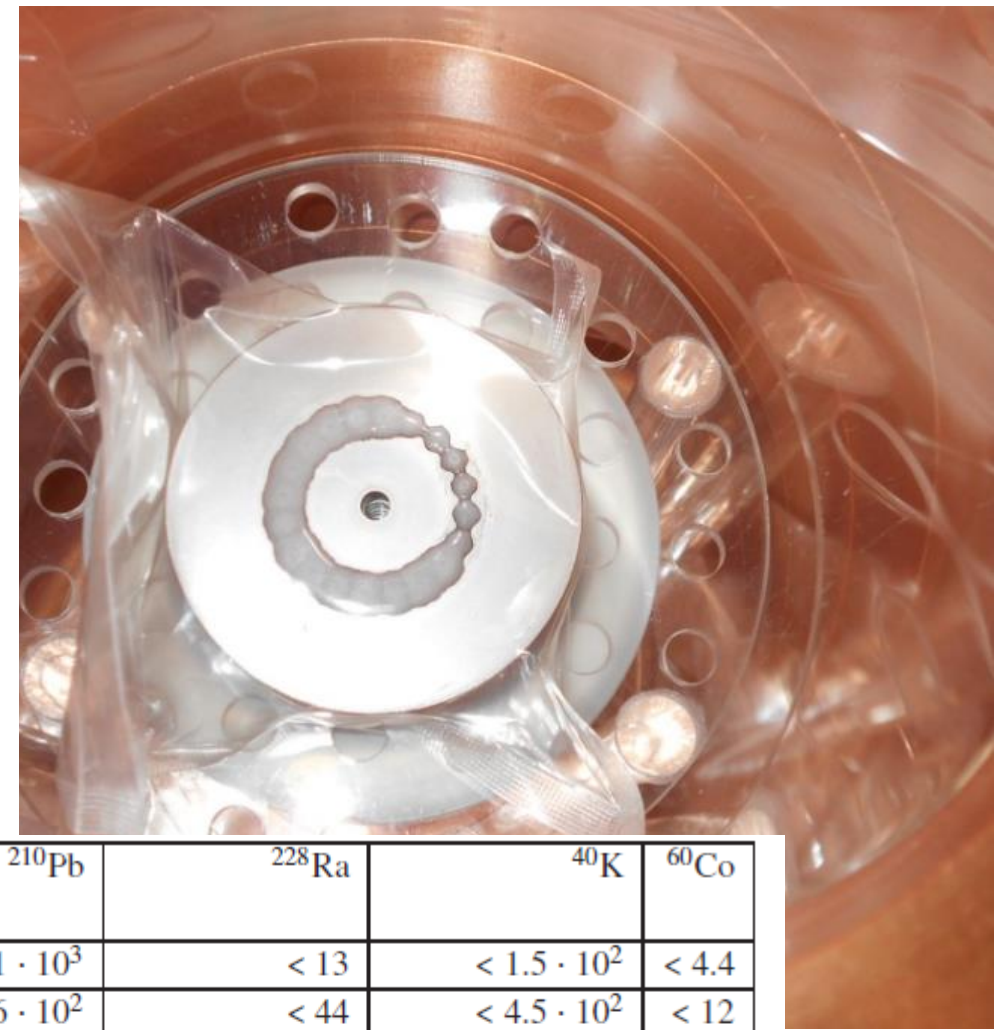
Samples	Materials	Weight for HPGe (g)	Weight per PMT (g)	Ratio
Body	Co free metal	4450	84.9	52.4
Stem	Co free metal	3557	62	57.4
Stem pin	Kovar alloy	92	1.6	58
Glass beads	Made from high-purity glass	186	2.3	80.8
Quartz window	Synthetic quartz	2610	82.1	31.8
Quartz dynode support	Synthetic quartz	360	11.8	30.5
Quartz spacer	Synthetic quartz	37.5	0.75	50
Al Seal	Aluminum	109.6	0.9	121.8
Kovar ring	Kovar alloy	196	3.3	59.4
Antimony for evaporation	Antimony	1.6	0.022	72.7
Al for evaporation	Aluminum	19.9	0.064	311
Hermetic	glass, Kovar	103	0.19	542
Lead wire, Ribbons	Nickel	189	1.88	101
Screw, nut, washer, getter stopper	Non-Magnetic (NM) stainless steel	115	1.2	95.8
Cathode	NM stainless steel (0.25 t)	133.5	2.4	55.6
Dynode 1st stage	NM stainless steel (0.3 t)	212	5.1	41.6
Dynode (2nd to 12th stage)	NM stainless steel (0.2 t)	375	7.5	50
Shield	NM stainless steel (0.3 t)	331	6.6	50.1
Spring, number plate	NM stainless steel	32.4	0.34	95.3
Anode mesh	NM stainless steel (0.25 t)	15.6	0.1	156
Disk stopper	NM stainless steel (0.18 t)	41.4	0.87	47.6
Dynode 1st stage mesh	SUS304 (0.05 t)	8.1	0.16	50.6
Disk	NM stainless steel (0.5 t)	654	12.6	51.9
Getter	Zr-Al alloy	7.0	0.07	100
Exhaust tube	Copper	86	1.4	60

- Measured as possible as large amount to increase sensitivity.
- Needed number is RI/PMT .
 - The important factor is how many PMTs the measured amount corresponds to.
 - Ratio listed
 - Amount for 30~500 PMTs were measured.



Glass beads

- Used as electrical insulation for the feedthroughs.
 - This type of insulation was used in low BG PMT from R10789.
- Though the amount is very small ~2g for each PMT, it has largest contribution for ^{226}Ra .
- To reduce RI, we synthesized it from high-purity reagents with the cooperation of the company.
 - With low RI reagents and also with low RI tools (milling tools).
- Resultant reduction
 - ^{226}Ra 810uBq/g \rightarrow <15uBq/g ($\sim 1/50$)
 - ^{40}K 1,400uBq/g \rightarrow 170uBq/g ($\sim 1/8$)

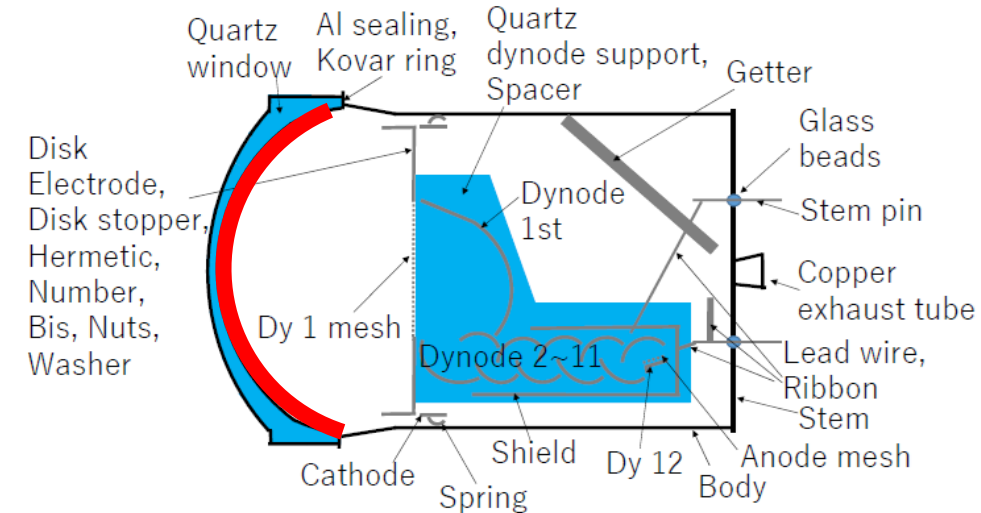


Samples	component ratio [%]	^{226}Ra	^{238}U (^{234}Th)	^{210}Pb	^{228}Ra	^{40}K	^{60}Co
SiO ₂ 5N grade	64	< 11	< $1.5 \cdot 10^2$	< $1.1 \cdot 10^3$	< 13	< $1.5 \cdot 10^2$	< 4.4
B ₂ O ₃ 4N5 Grade	3	< 45	< $1.8 \cdot 10^2$	< $4.6 \cdot 10^2$	< 44	< $4.5 \cdot 10^2$	< 12
Na ₂ B ₄ O ₇ 4N5 Grade	24	$(1.8 \pm 0.6) \cdot 10^2$	< $3.9 \cdot 10^2$	< $1.3 \cdot 10^3$	< 74	< $1.1 \cdot 10^3$	< 25
Al(OH) ₃ 4N grade	9	< 25	< $2.3 \cdot 10^2$	< $1.5 \cdot 10^3$	< 42	< $2.0 \cdot 10^2$	< 7.6
Sum		36 ± 15	< $1.3 \cdot 10^2$	< $6.5 \cdot 10^2$	< 19	< $2.6 \cdot 10^2$	< 6.6
Glass after synthesis		12 ± 4	57 ± 26	< 61	< 9.4	$(1.5 \pm 0.5) \cdot 10^2$	< 2.5
Glass after milling		24 ± 5	< 97	< 99	17 ± 4	$(1.3 \pm 0.6) \cdot 10^2$	< 4.0
R13111 Glass beads		< 15	< 72	< $1.9 \cdot 10^2$	< 9.7	$(1.7 \pm 0.7) \cdot 10^2$	< 2.0
R10789 R10789 glass beads		$(8.1 \pm 0.1) \cdot 10^2$	$(3.3 \pm 0.6) \cdot 10^2$	< $4.1 \cdot 10^2$	$(1.4 \pm 0.1) \cdot 10^2$	$(1.4 \pm 0.1) \cdot 10^3$	< 2.3

uBq/g

Photo cathode

- Photo cathode
 - Most of the 40K in R10789 is come from the photocathode material.
 - Chemicals used to make photocathode material contain potassium, which contain 40K with a natural abundance of 117 ppm.
 - We cannot remove potassium, so 39K enriched material was used to make the photocathode.
 - 40K in a material reduced to $\sim 1/100$.

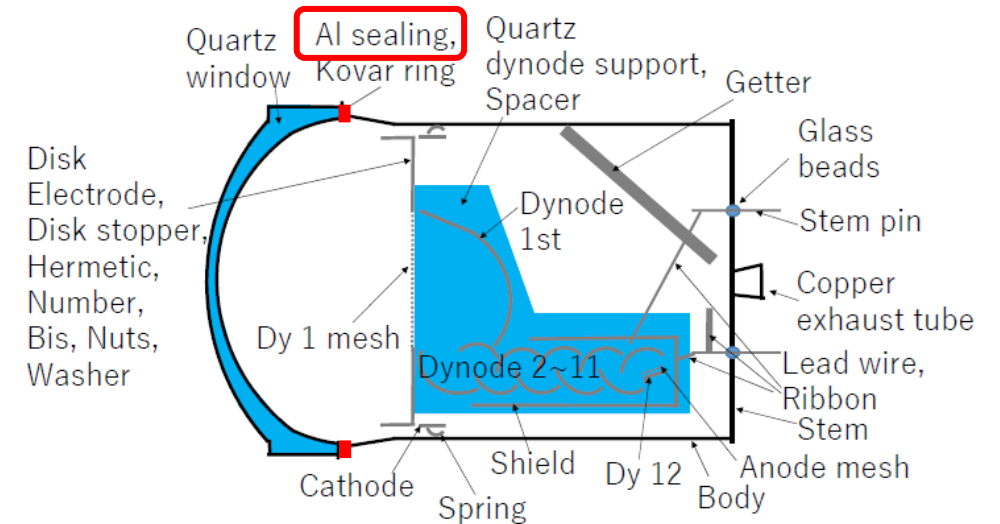


Samples	^{226}Ra	^{238}U (^{234}Th)	^{210}Pb	^{228}Ra	^{40}K	^{60}Co
without enrichment	< 82	< $3.0 \cdot 10^4$	< $8.8 \cdot 10^4$	< $3.4 \cdot 10^2$	$(1.0 \pm 0.1) \cdot 10^7$	< $9.9 \cdot 10^2$
with enrichment	< $2.2 \cdot 10^2$	< $1.3 \cdot 10^3$	< $8.9 \cdot 10^3$	< $1.9 \cdot 10^2$	$(8.4 \pm 0.4) \cdot 10^4$	< 52

uBq/g HPGc measurement with 20g material

Aluminum seal

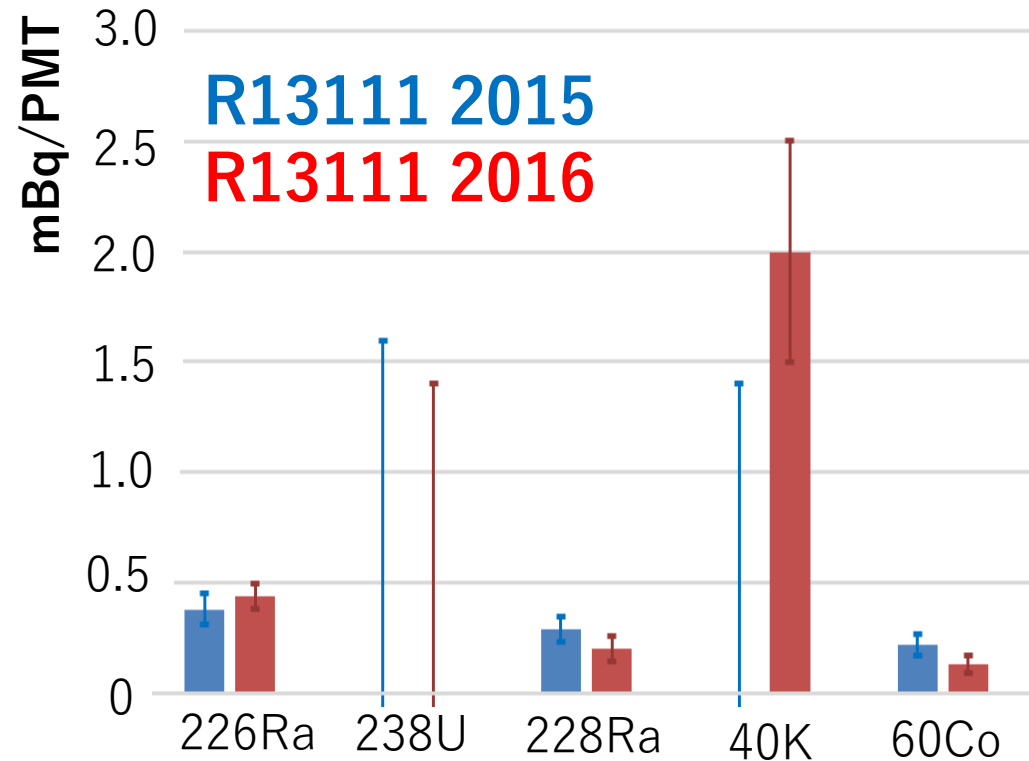
- Aluminum seal
 - Largest ^{238}U contributor.
 - Sealing between the quartz window and body. This position makes this material as large BG source in the XMASS-I.
 - Higher purity material was used for R13111, changed from 98.5% for R10789 to 5N for R13111.
- $< \sim 1/100$



Samples	^{226}Ra	^{238}U	^{210}Pb	^{228}Ra	^{40}K	^{60}Co	^{238}U	^{232}Th	Method for ^{238}U and ^{232}Th
used in 2015 Production (5N purity)	< 10	< 51	$< 1.0 \cdot 10^2$	13 ± 5	$< 1.8 \cdot 10^2$	< 5.8	2.5	$4.1 \cdot 10^{-1}$	GD-MS
used in 2016 Production (5N purity)	< 2.8	< 37	< 66	< 2.5	< 34	$< 9.6 \cdot 10^{-1}$	2.5	$4.1 \cdot 10^{-1}$	GD-MS
used in R10789 PMT (98.5% purity)	< 36	$(3.3 \pm 0.4) \cdot 10^3$	$(7.7 \pm 6.0) \cdot 10^3$	$(1.0 \pm 0.3) \cdot 10^2$	$< 7.1 \cdot 10^2$	< 18	$3.9 \cdot 10^3$	74	ICP-MS

uBq/g

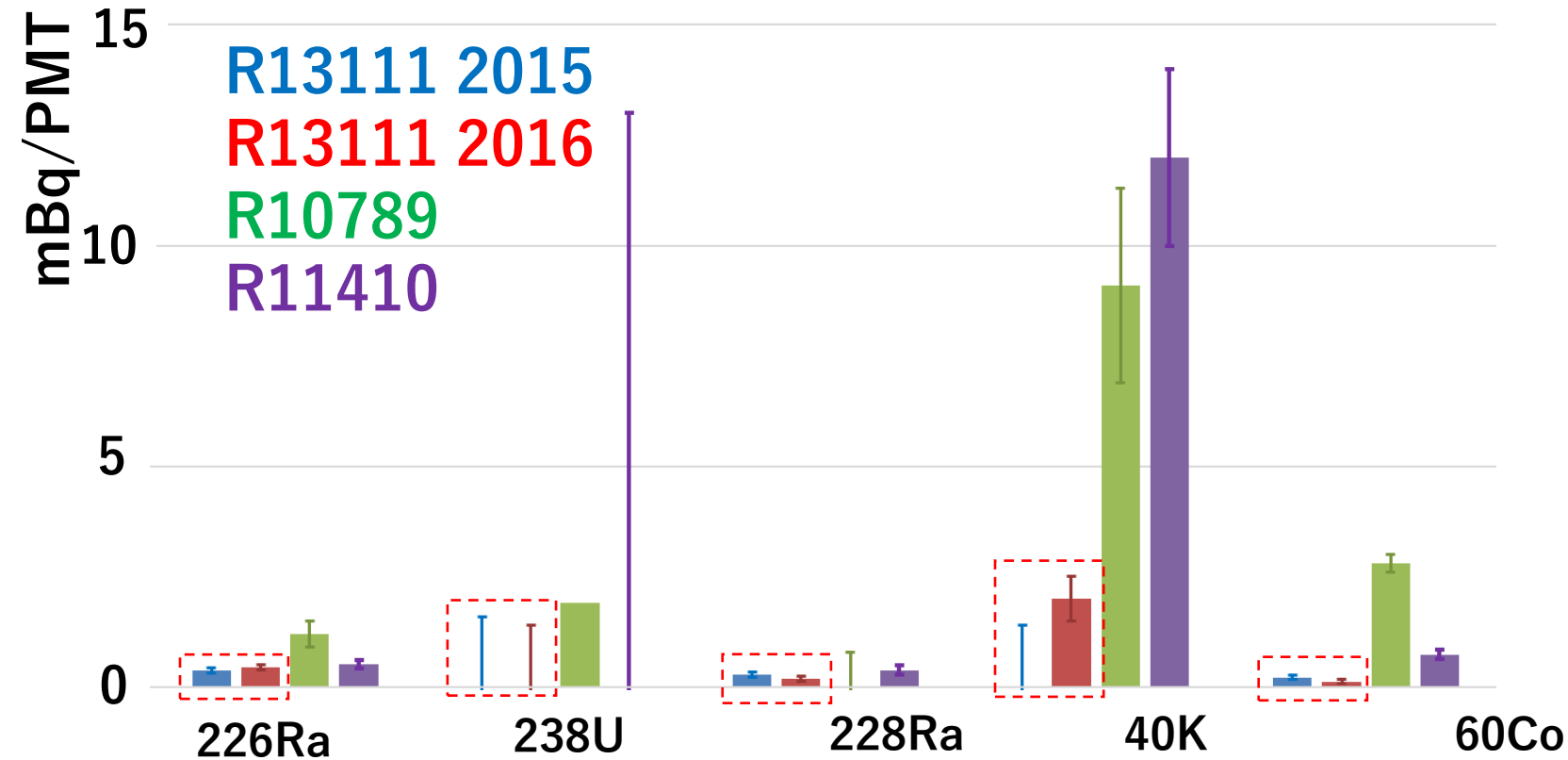
RI of developed R13111



- RI for assembled PMT
- By measuring 13 PMTs in one measurement, achieved high sensitivity.
 - 226Ra ~0.4mBq/PMT
 - 238U <1.6mBq/PMT
 - 228Ra 0.3mBq/PMT
 - 40K ~2mBq/PMT
 - 60Co ~0.2mBq/PMT
- Two production lots at 2015 and at 2016 version uses same parts.
 - Consistent results.
 - We can control difference due to different lot in this error level.

[mBq/PMT]	226Ra	238U	228Ra	40K	60Co
R13111(2015)	0.38 ± 0.07	<1.6	0.29 ± 0.06	<1.4	0.22 ± 0.05
R13111(2016)	0.44 ± 0.06	<1.4	0.2 ± 0.06	2.0 ± 0.5	0.13 ± 0.04

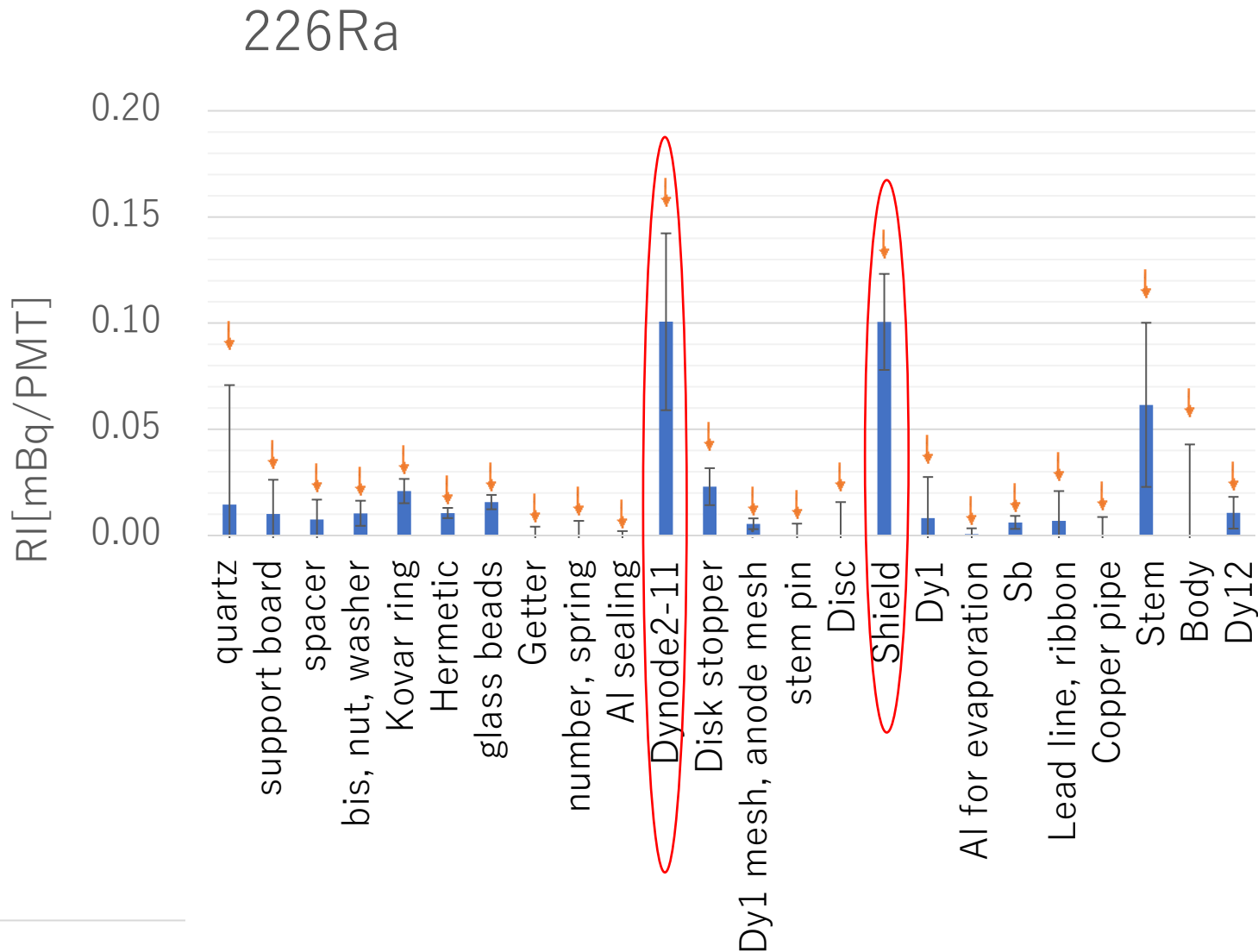
Comparison to Other PMTs



- Large reduction from R10789
 - ^{226}Ra $\sim 1/3$
 - ^{228}Ra $\sim 1/3$
 - ^{40}K $\sim 1/5$
 - ^{60}Co $\sim 1/10$
- Sensitive area difference is not considered.
 - R10789 2 inch
 - R13111 3 inch
 - Another factor of $\sim 1/2$

[mBq/PMT]	226Ra	238U	228Ra	40K	60Co
R13111(2015)	0.38 ± 0.07	<1.6	0.29 ± 0.06	<1.4	0.22 ± 0.05
R13111(2016)	0.44 ± 0.06	<1.4	0.2 ± 0.06	2.0 ± 0.5	0.13 ± 0.04
R10789	1.2 ± 0.3	1.9 (Parts sum)	<0.78	9.1 ± 2.2	2.8 ± 0.2
R11410-21 (XENON1T)	0.52 ± 1.0	<13	0.39 ± 0.1	12 ± 0.2	0.74 ± 0.1

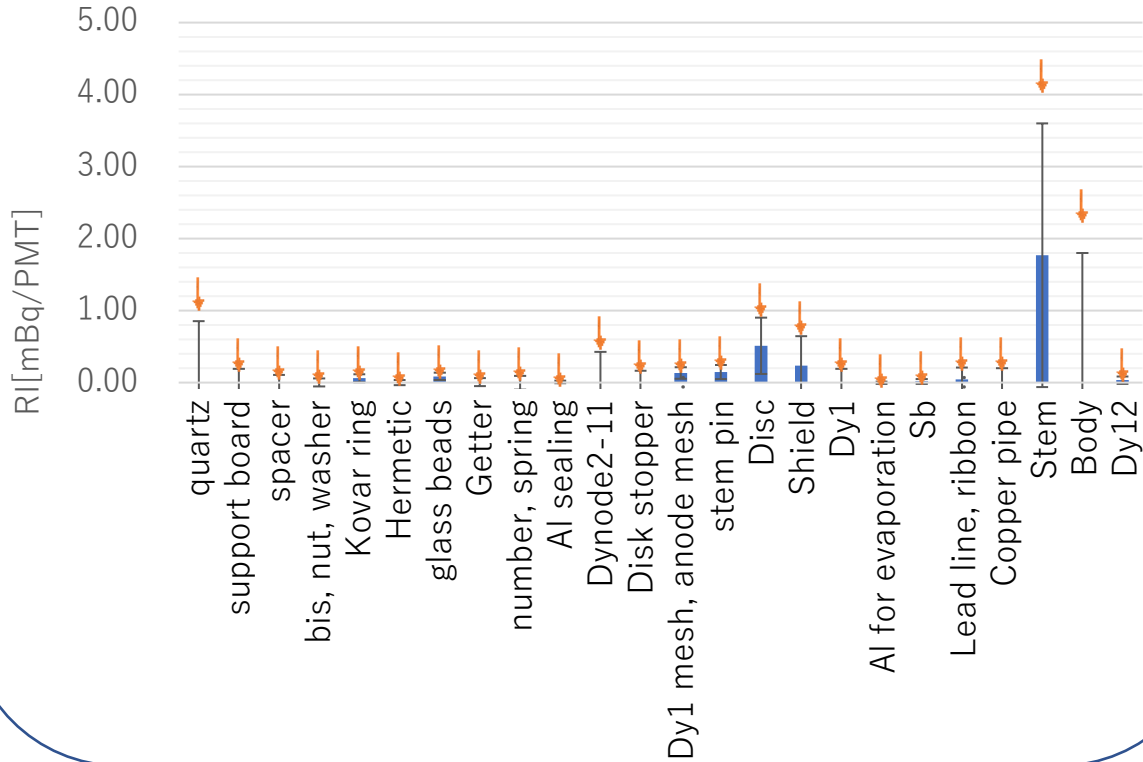
Contribution from each component



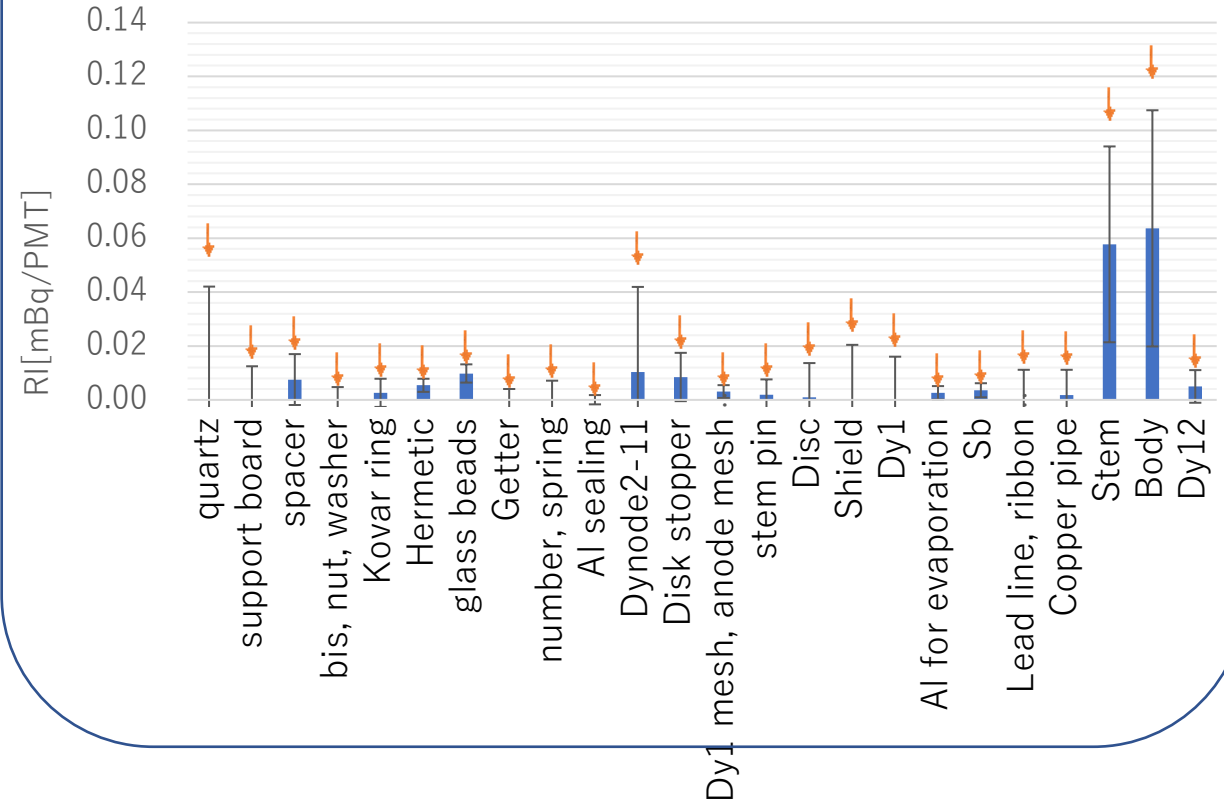
- 226Ra
- Basically error becomes small for component whose amount used for one PMT is small. We can measure large amount at one measurement.
- Contributions from glass beads reduced largely.
 - 1.6mBq/PMT(R10789)
->0.016mBq/PMT
- Some stainless steel parts have large contributions,
 - Dynodes and shield.
 - Stem's contribution is not significant, it has large error.

Contribution from each component

238U

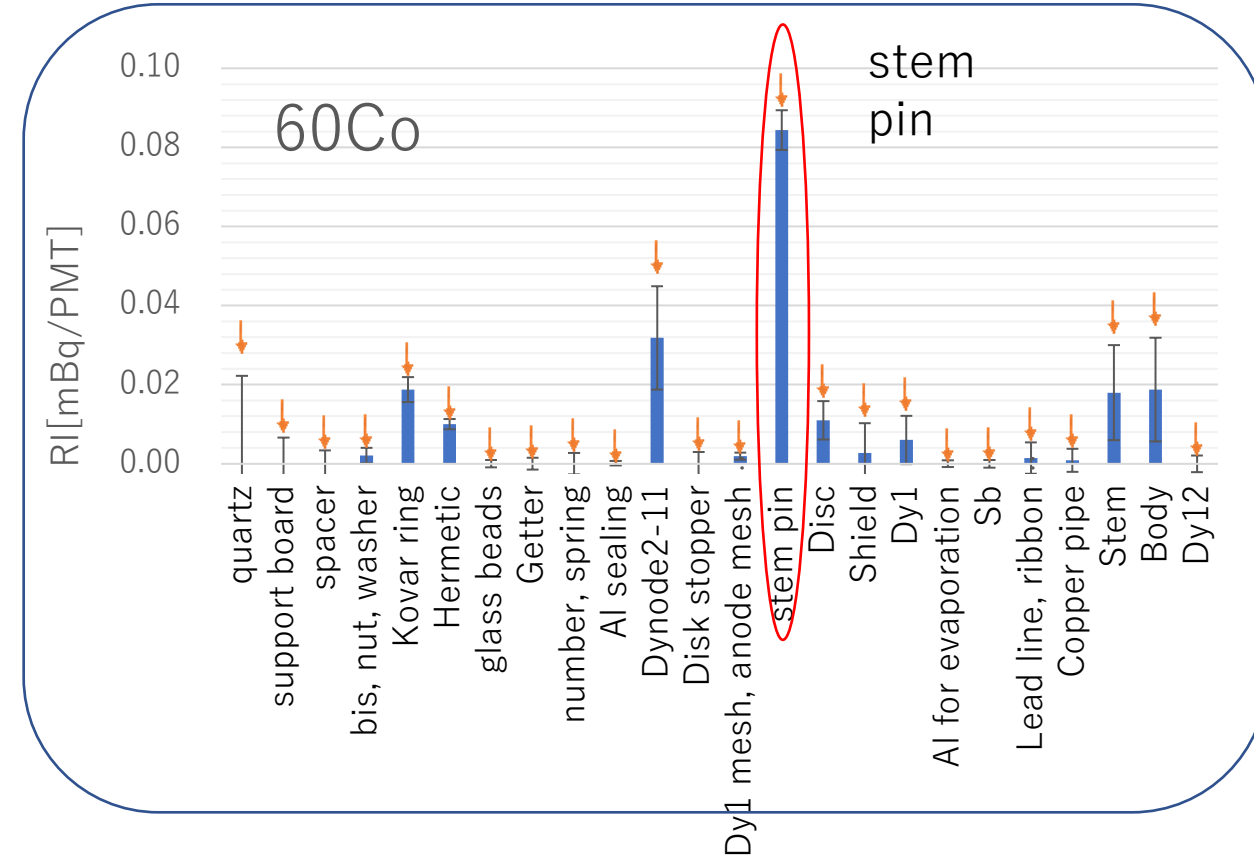
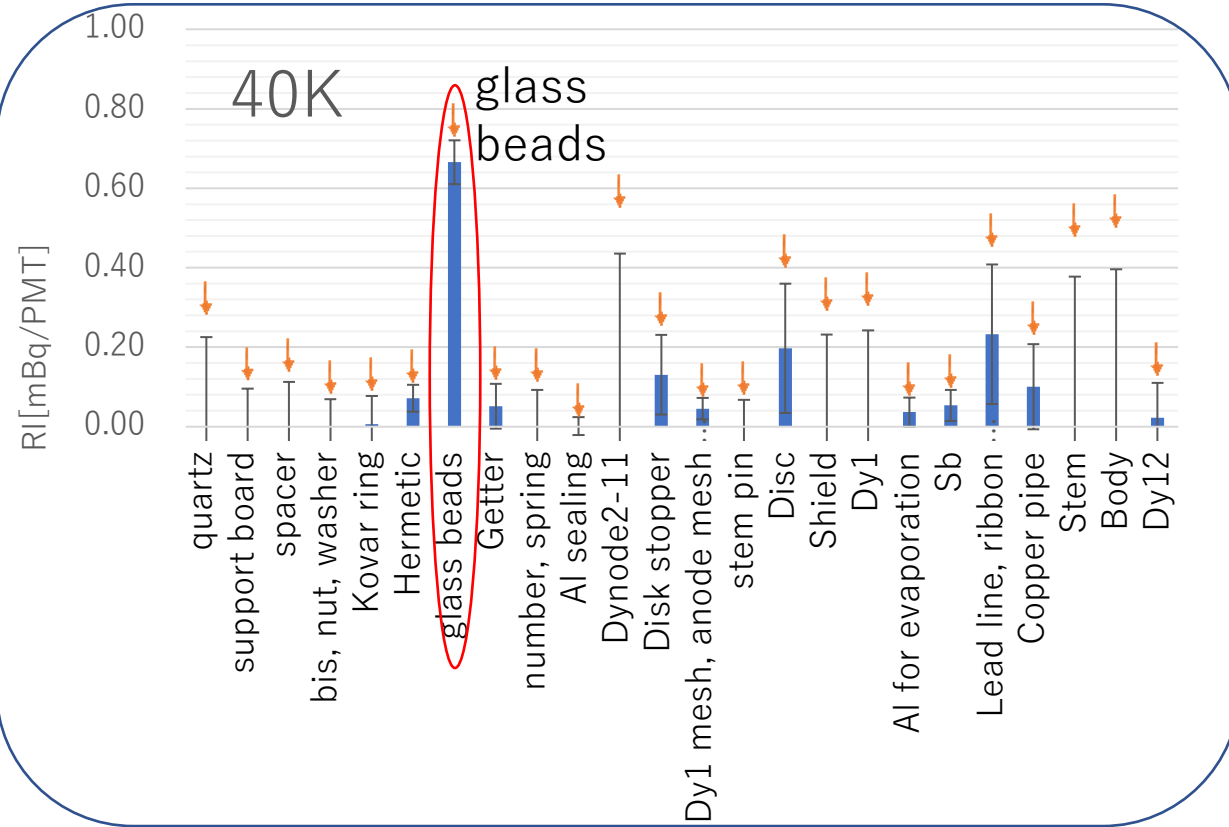


228Ra



- Contributions from glass beads and Al sealing became negligible.
- 238U and 228Ra, no significant contributions were identified.

Contribution from each component



- 40K

- Glass beads contribution reduced from $\sim 3\text{mBq/PMT}$ (R10789) $\rightarrow 0.6\text{mBq/PMT}$.
- Though still significant and major contribution.

- 60Co

- Body and stem changed from Kovar metal to Co free metal.
- Largest contribution comes from stem pin made from Kovar.

RI reduction

- **Achieved Large reduction**
 - The lowest level available at present
- For further reduction
 - Stainless steel
 - Glass
 - Kovar
 - Higher sensitivity of measurement

[mBq/PMT]	226Ra	238U	228Ra	40K	60Co
R13111(2015) 3 inch	0.38 ± 0.07	<1.6	0.29 ± 0.06	<1.4	0.22 ± 0.05
R13111(2016) 3 inch	0.44 ± 0.06	<1.4	0.2 ± 0.06	2.0 ± 0.5	0.13 ± 0.04
R10798 2 inch	1.2 ± 0.3	1.9 (Parts sum)	<0.78	9.1 ± 2.2	2.8 ± 0.2
R11410-21 3 inch (XENON1T)	0.52 ± 1.0	<13	0.39 ± 0.1	12 ± 0.2	0.74 ± 0.1
R12699 v1 2 inch	0.45 ± 0.11	<1.03	<0.40	30.83 ± 2.14	<0.07

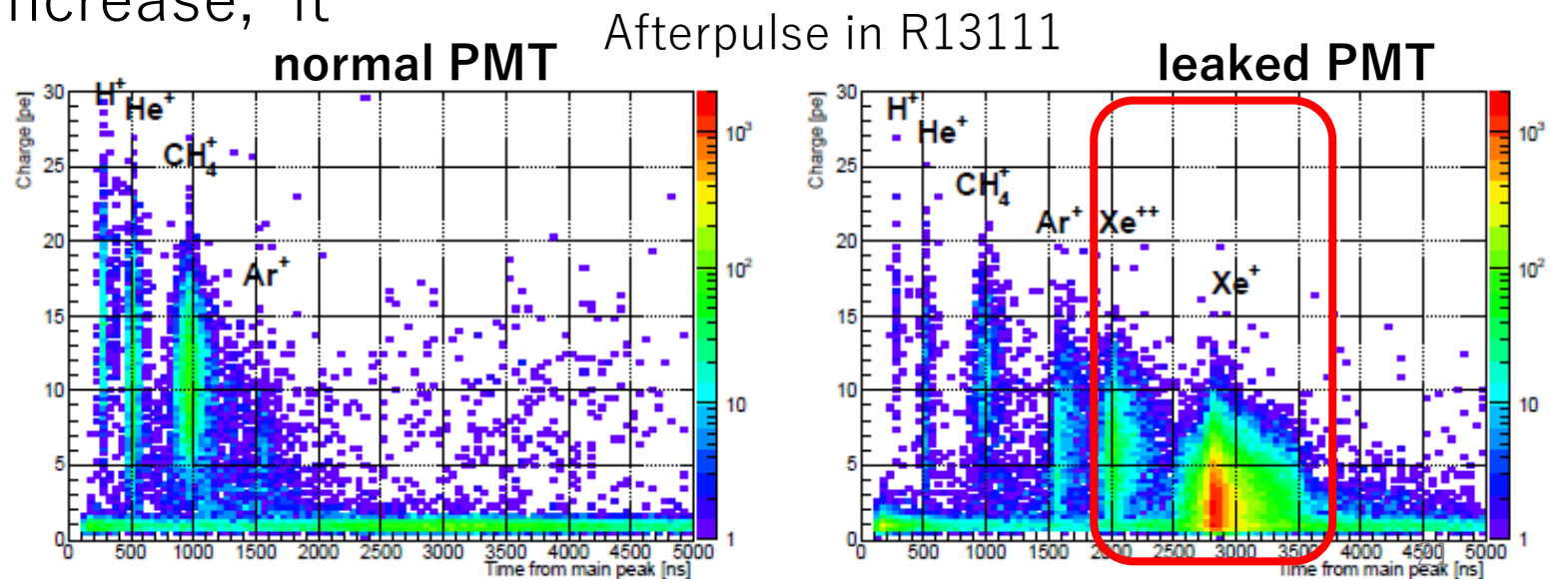
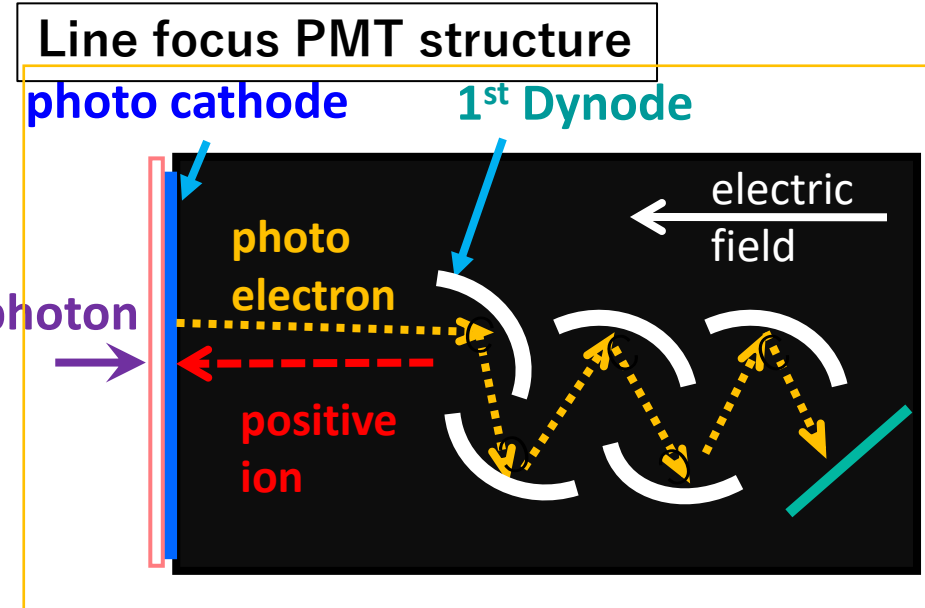
Eur. Phys. J. C **75** (2015) 546 [arXiv:1503.07698[astro-ph.IM]]

“Low background control for PandaX” by Ke Han at TAUP2023

Study of Xe leak problem

Study of Xenon leak problem

- We found that many R13111 shows Xe after pulse
 - $\sim 1/3$ (16 in 58)
 - Xe leak inside PMT
 - Ionized residual gas inside PMT makes after pulse
 - Large BG source
 - If leak continues to increase, it becomes impossible to apply HV.
- We studied cause and countermeasure.

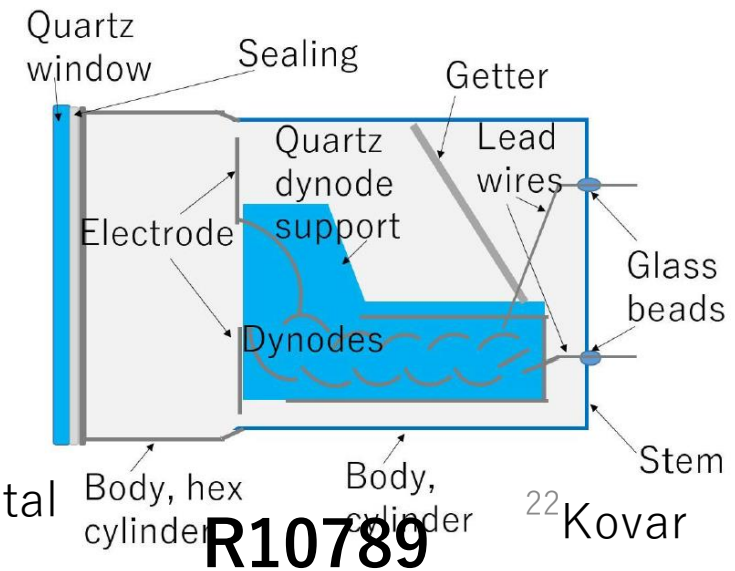
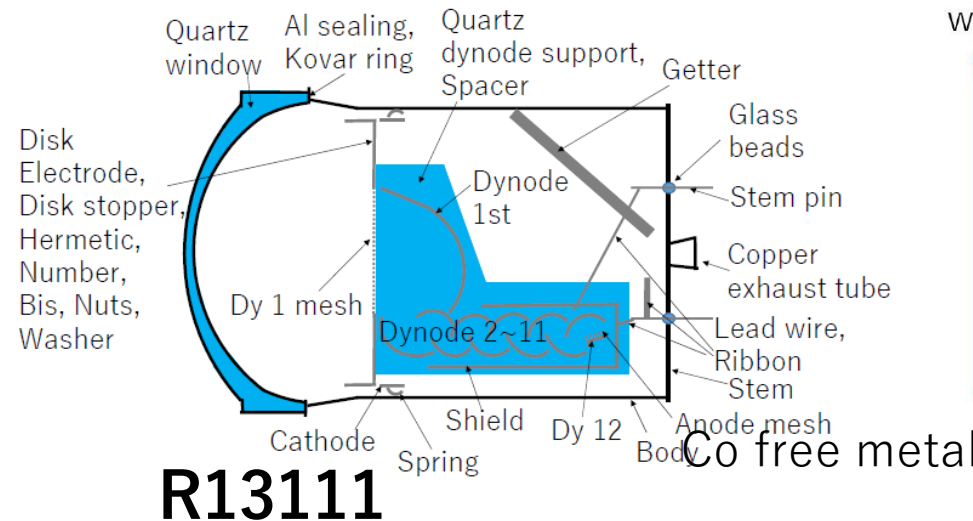
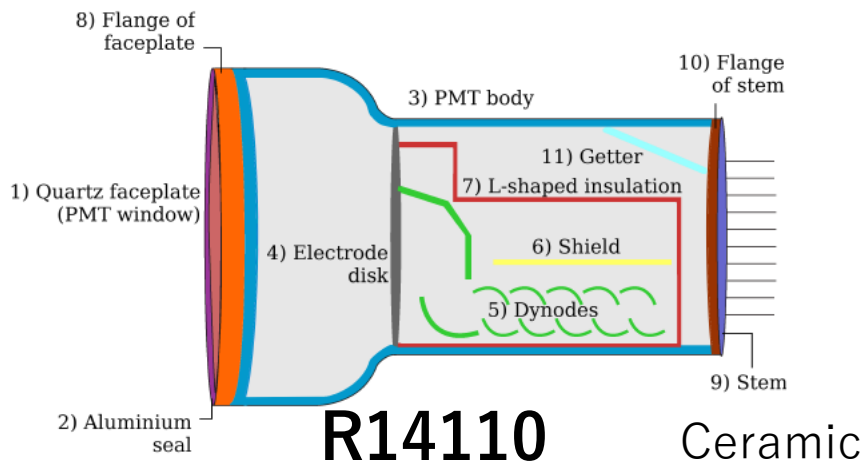


Identification of the cause

- Xe leaks in other similar PMTs

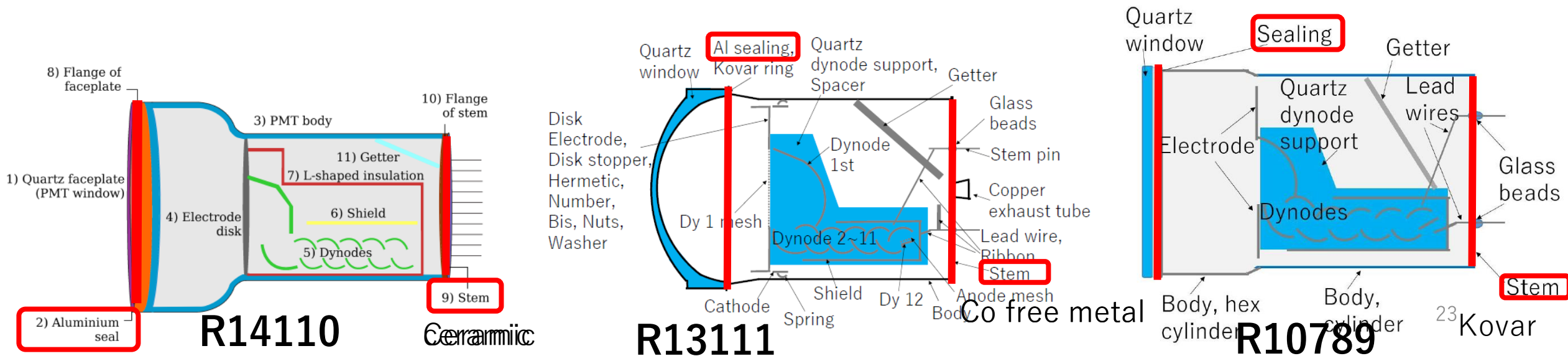
- R11410 cfJINST 12 P01024 (2017) XENON1T PMT paper
 - similar Xe after pulse
 - 8 in 44
 - many structures are common with R13111 and R10789.
- R10789
 - no such after pulse
 - 0 in 642 during XMASS-I operation

- The structure causing the leak is common with R11410 but different from R10789.



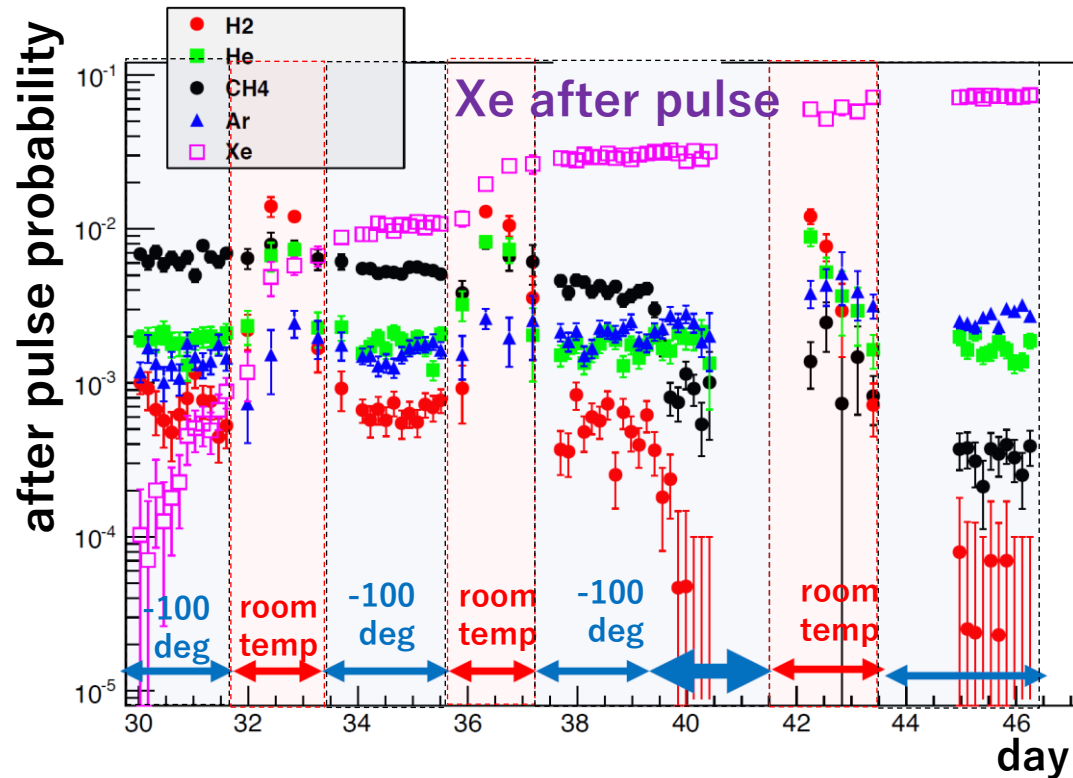
Identification of the cause

- Different from R10789 and Common with R11410
- 5N aluminum seal
- It was found that the seal surface of the R13111 PMT had many tiny scratches compared to that of the R10789 PMT by microscope study.
- To confirm, we prepared PMTs with two different aluminum seals.
 - 23 R11410 with 4N Al seal
 - 28 R11410 with 5N Al with special care
 - Applied cooling cycles inside Xe



Leak check with different Al seal

Example of Xe leak during cooling cycle



- Check after pulse probability
 - Probability for one single pe signal by LED.
- For many cases Xe leak increased with every cooling cycle.
 - All first leaks were made during 1st or 2nd cycle.
 - All exceed 0.1% probability
 - We named this “large leaks”
- In some cases leak were not increased.
 - Probability for those PMT were not exceed 0.1%
 - named this “small leaks”

Study results

	Tested number	Large leaking	Small leaking	Large leak probability
R11410 5N Al cfJINST 12 P01024 (2017) XENON1T PMT paper	44	7	1	16+/-7%
R11410 4N Al	23	0	0	<10%
R11410 5N Al with special handling	28	0	0	<8%

- Large leaking
 - R11410 5N 16+/-7%
 - R11410 4N <10% (90%CL)
 - R11410 5N with special handling <8% (90%CL)
- We confirmed that
 - 5N aluminum seal is the cause (at least one of the main causes) of Xe leak.
 - Surface status of seal is the matter
 - Exchange this 5N to 4N Al or 5N but with special care can solve the problem.

Summary

- PMT is one candidates for future low BG experiments.
- Ultra low background PMT R13111
 - Developed for future rare event search experiments
- Achieved RIs are
 - ^{226}Ra $\sim 0.4\text{mBq/PMT}$
 - ^{238}U $< 1.6\text{mBq/PMT}$
 - ^{228}Ra 0.3mBq/PMT
 - ^{40}K $\sim 2\text{mBq/PMT}$
 - ^{60}Co $\sim 0.2\text{mBq/PMT}$
 - the lowest level available at present
- Xe leak problem
 - Cause was the 5N Al seal
 - 4N Al or 5N but with special care can solve the problem.