



Rn assay and distillation purification of Lq. Xe for Kr

Yasuo Takeuchi

(Kamioka Observatory, ICRR, Univ. of Tokyo)

- Introduction
- Distillation purification of Xe for Kr
- Kr assay with API-MS detector
- Rn assay with XMASS prototype detector
- Summary

Why liquid xenon scintillator

Scintillation light	~42photon/keV
Scintillation light wave length	175nm
Scintillation light width	~40nsec
Atomic number	54
Atomic weight	131.29 amu
Density	3.0 g/cm ³
Melting (boiling) point	161.4K (165.1K)
Chemical series	Noble gases

- High photon yield
 - Low threshold, good energy resolution, ...
- Can be directly read by PMT
- Large atomic number
 - Radiation length ~2.4cm
 - **Self shielding against external backgrounds**
 - Compact (R=1.22m for 23 tons)
- Easy to liquefy
 - Liquid N₂ can be used
- Various purification method
 - **Distillation**, circulation during experiment, ...
 - **Effective reduction against internal backgrounds**
- No long life radioactive isotopes
- ¹³⁶Xe is a ββ decay candidate

Internal backgrounds

■ ^{238}U series

- ^{222}Rn ($\tau_{1/2} = 3.8\text{d}$, 3.3MeV beta), ...
- Target (for XMASS 800kg detector): $1 \times 10^{-14}\text{g}(^{238}\text{U})/\text{g}(\text{Xe})$

■ ^{232}Th series

- ^{220}Rn (55s, 2.3MeV beta[64%]), ...
- Target: $2 \times 10^{-14}\text{g}(^{232}\text{Th})/\text{g}(\text{Xe})$

■ ^{85}Kr

- Contamination during manufacture and refinement
- $\tau_{1/2} = 10.8\text{y}$, 687keV beta (99.6%)
- Target: Kr = 1 ppt (mol) (assuming $^{85}\text{Kr}/\text{Kr} = 1.2 \times 10^{-11}$)

(~ppb level Kr for commercial “Kr-free” Xe)



Distillation purification of Xe for Kr

Distillation of Xe for Kr

Distillation tower

Distillation system in Kamioka

Distillation of Xe for Kr

■ Impurities in xenon

- CO₂, H₂O : removed by adsorption
- Kr, O₂, N₂, H₂, He : removed by **distillation**
(boiling points are lower than Xe)

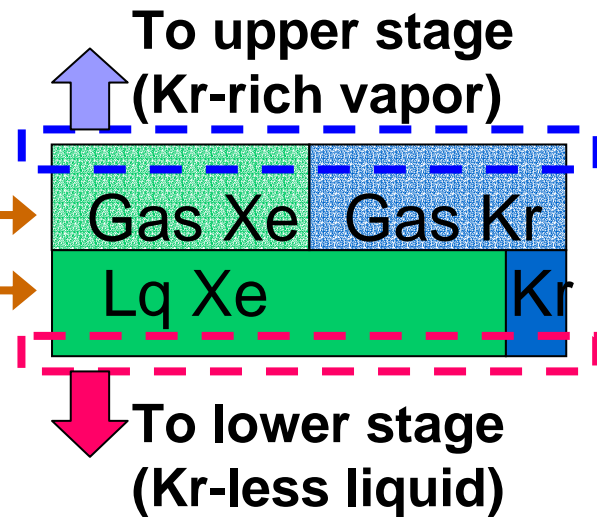
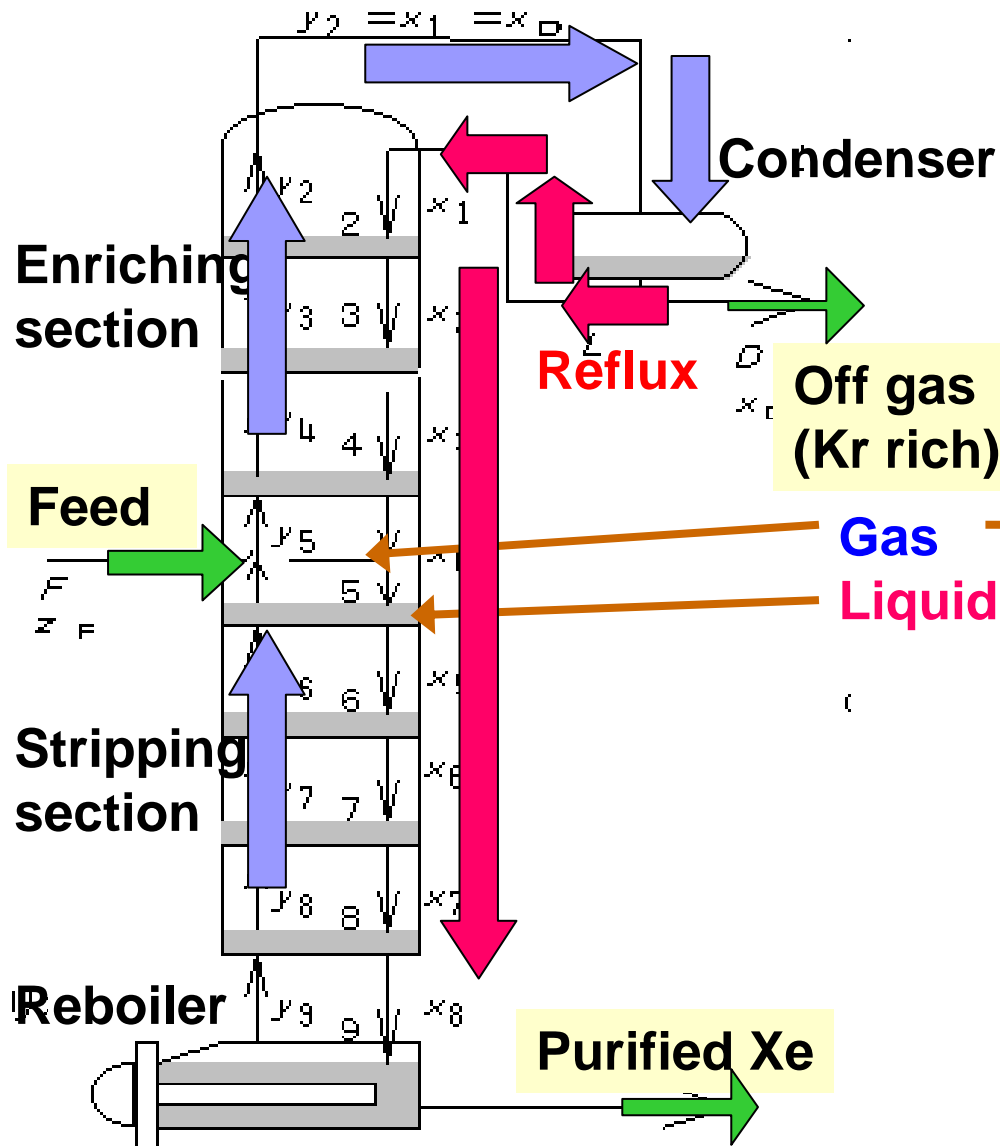
	Boiling point (@1 atm)	Boiling point (@2 atm)
Xe	165K	178K
Kr	120K	129K

■ Built a Xe distillation system in Kamioka mine

- Process speed: **0.6kg/hour** (= ~100kg / 1 week)
- **99% yield** (= 99% Kr-less gas, 1% Kr-rich gas)
- **1/1000 Kr** in purified Xe (design value)
- Operation condition: **178~180K, 2atm** (measured)

Distillation tower

- Multiple stages in a tower
- Each stage is in vapor-liquid equilibrium
- **Volatilities** are different between Xe and Kr



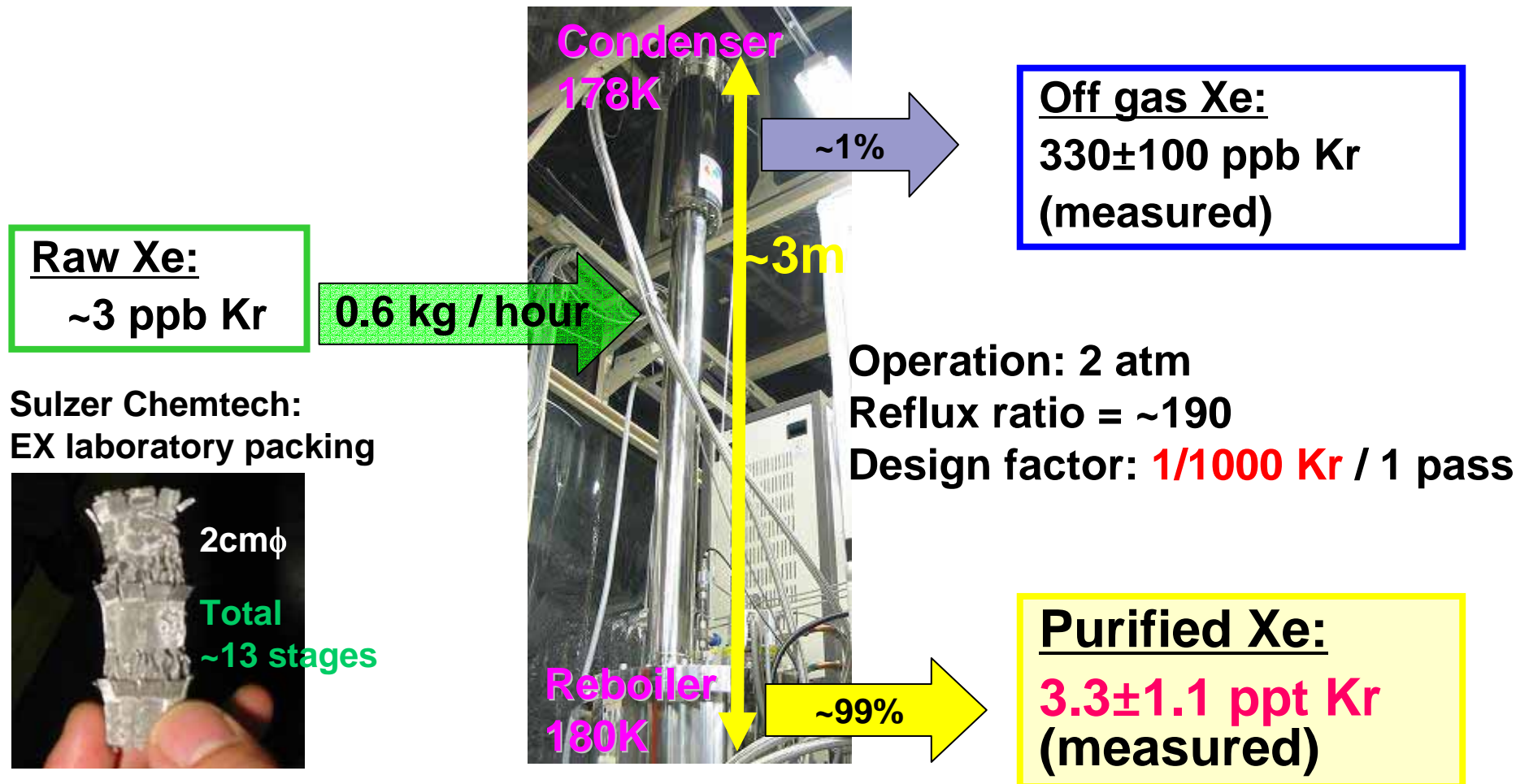
Relative volatility

$$= \frac{(\text{Kr in gas} / \text{Kr in lq})}{(\text{Xe in gas} / \text{Xe in lq})}$$

$$= \mathbf{10.4} \text{ (@178K)}$$

Distillation system in Kamioka

- A special distillation system of Xe for Kr was built in Kamioka Observatory
- We have processed 100kg Xe in March '04



December 13, 2004

Y.Takeuchi @LRT2004 in Sudbury

Kr assay with API-MS detector

GC + API-MS system

Measurement of Kr in purified Xe

Kr assay result

GC + API-MS system

- API: **A**tmospheric **P**ressure **I**onization
- Primary ionization by **Corona discharge**
 - Small fraction of carrier and target molecules are ionized
- **Secondary ionization by ion-molecule reaction**
 - Carrier gas (C): higher ionization potential
 - Target (X): lower ionization potential

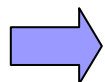


(Most of target molecules are ionized by charge exchange)

- Ionization potential:

	He	Ar	Kr	Xe
eV	24.6	15.8	14.0	12.3

- **Need to extract Kr from Xe**



Gas Chromatography (GC) + API-MS system

GC + API-MS system at SAAN

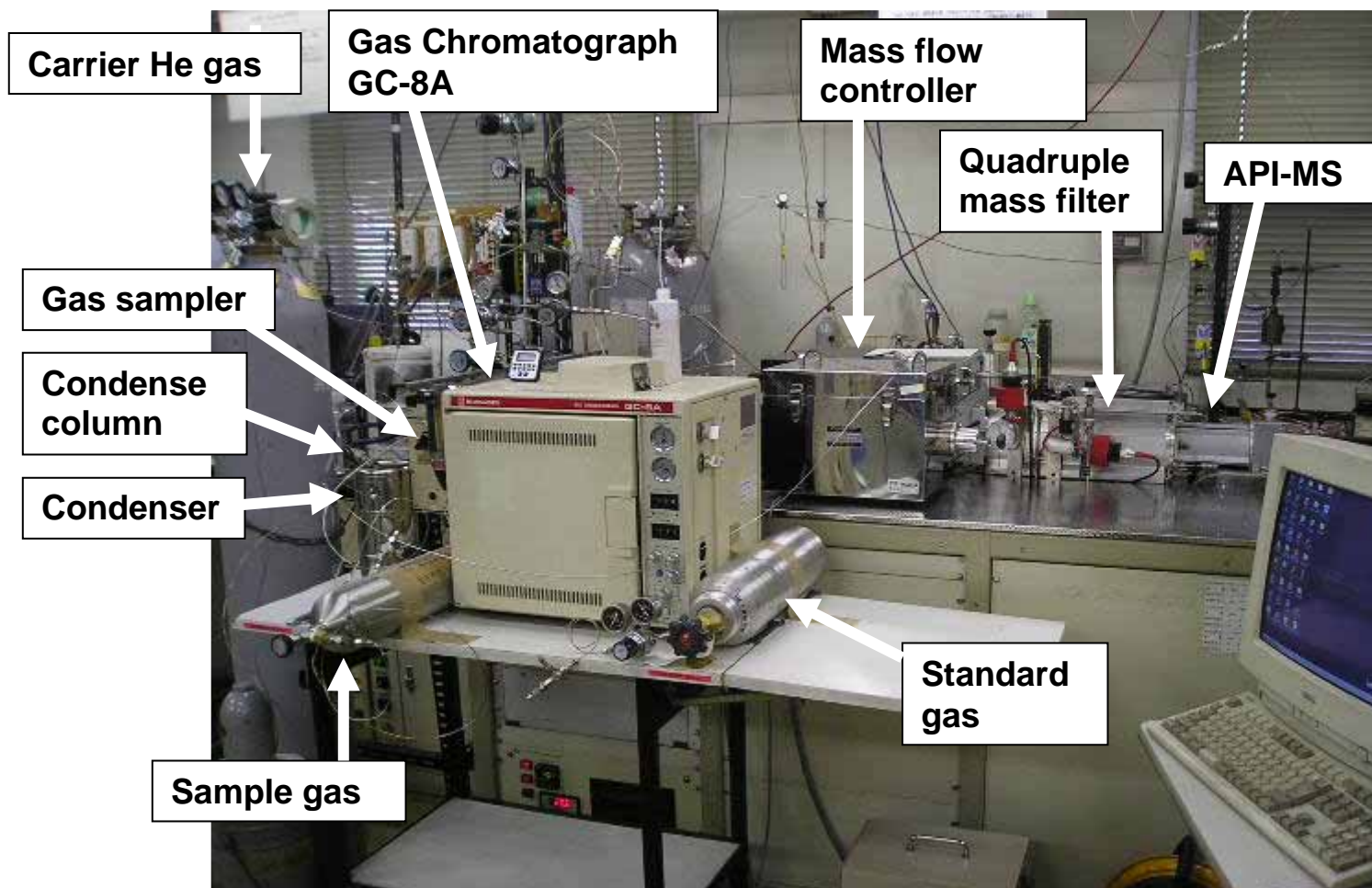
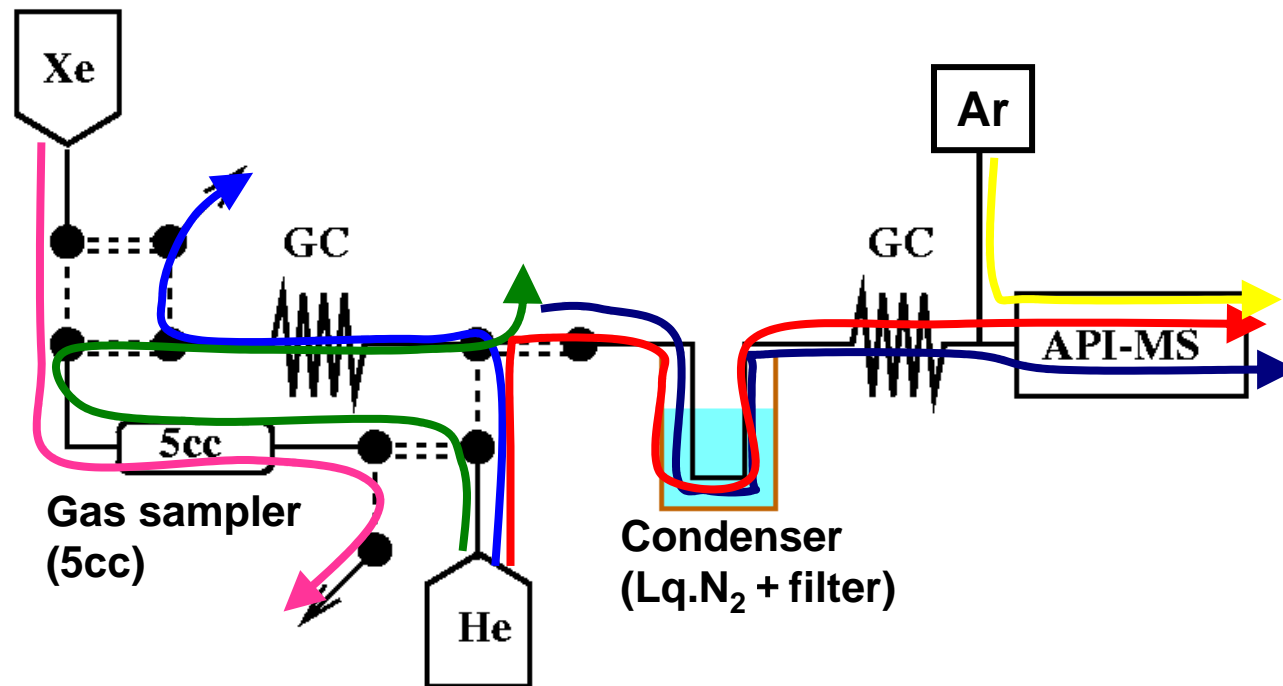


Photo by Kawaguchi Laboratory,
Research & Development Division,
Taiyo Toyo Sanso Co. Ltd.

Measurement of Kr in purified Xe

- Extract Kr from Xe by GC with He carrier gas, then trap Kr in the condenser. Repeat this 100 times.
- Feed the trapped Kr into API-MS with He and Ar carrier gas

~15min / cycle



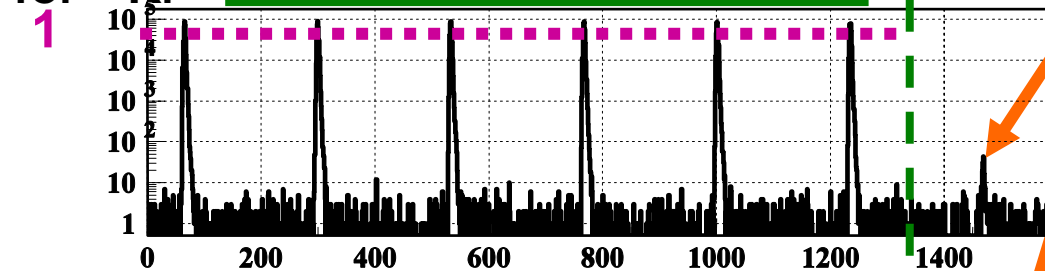
- Purge GC with He
- Sample 5cc Xe
- Feed sample Xe into GC with He carrier
- Only Kr timing, feed sample gas into the condenser.
- Repeat above
- After heat up, feed condensed Kr into API-MS (select only Kr timing by GC)

Kr assay result

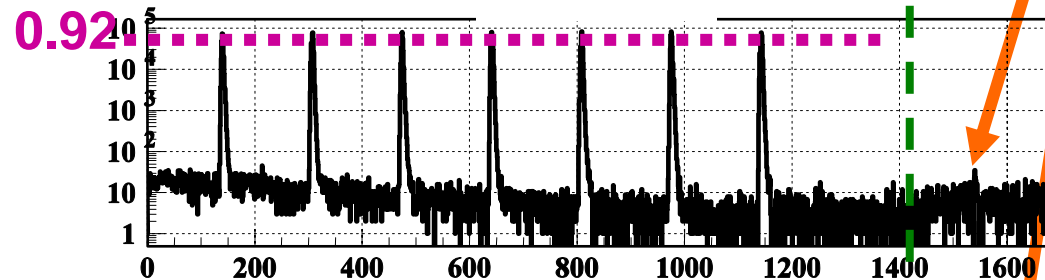
API-MS
count
for ^{84}Kr

Calibration with
1ppm Kr standard gas

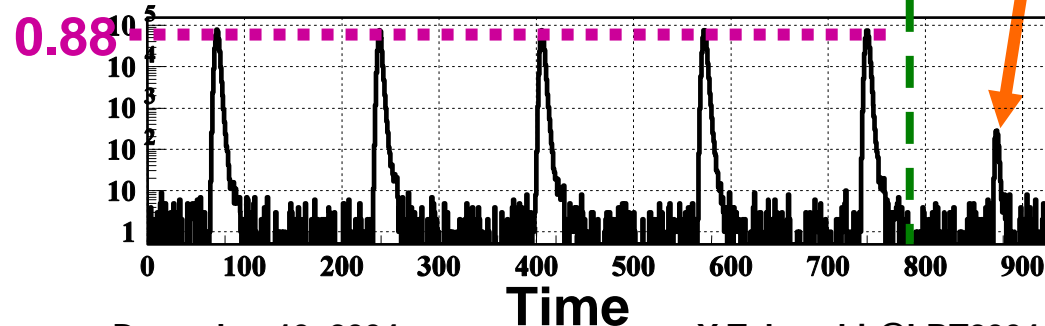
Measurement after 100
times condense



Sample Xe gas 184.0 ± 15.8 count
Carrier He gas $(88.5 \pm 23.9) / 0.92$
Kr 50ppt $(1176.9 \pm 35.7) / 0.88$



Kr concentration =
(Sample Xe - He gas)
/ Kr50ppt gas \times 50ppt
= 3.3 ± 1.1 ppt (stat. error only)



Factor $\sim 1/1000$ / 1pass
was achieved



Rn assay with XMASS prototype detector

Strategy of the XMASS project

XMASS prototype detector

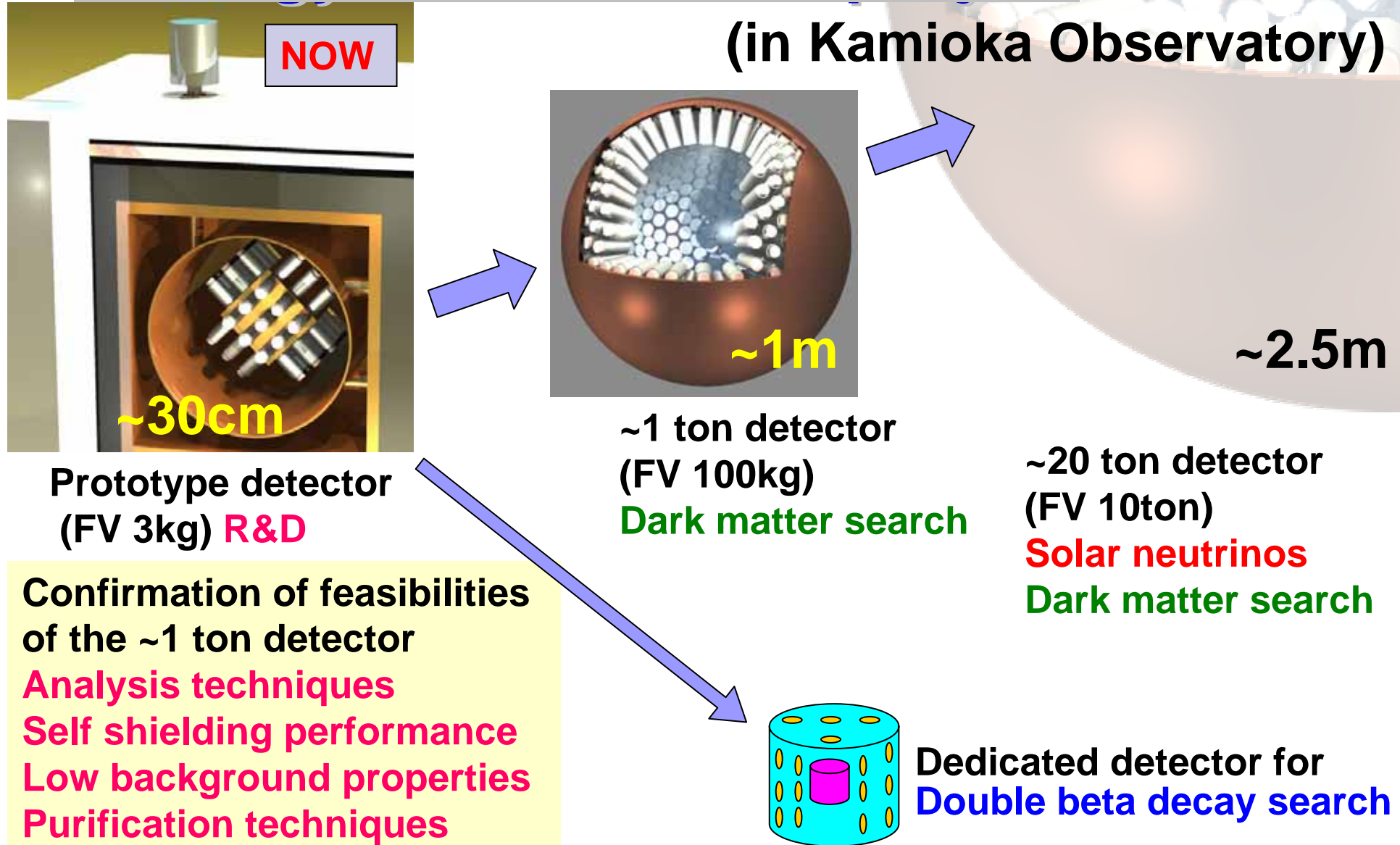
Rn assay with prototype detector

^{222}Rn measurements

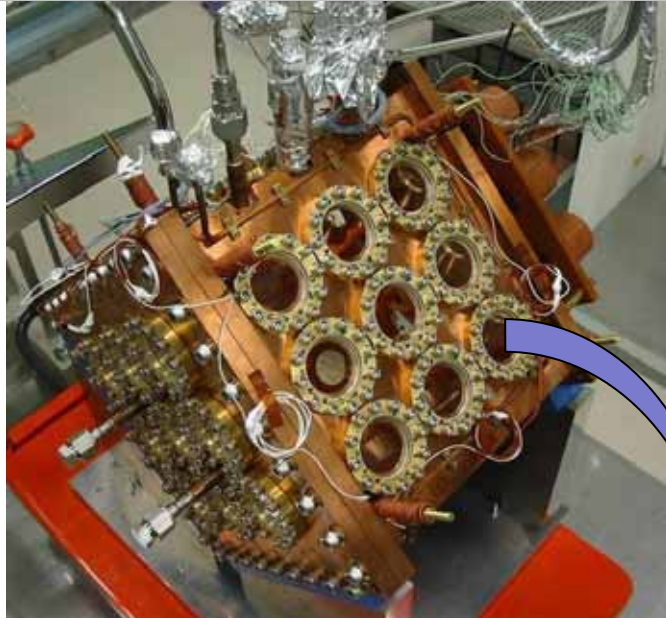
Internal background sources

Strategy of the XMASS project

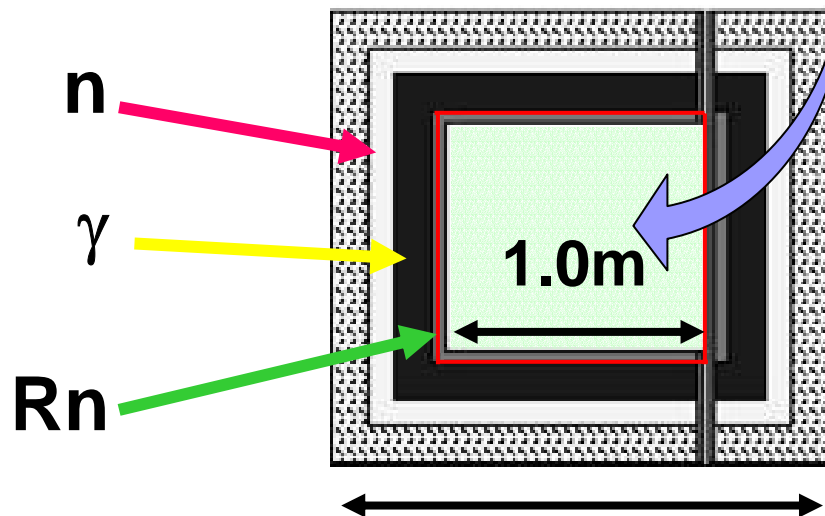
(in Kamioka Observatory)



XMASS prototype detector



- 30 liter liquid Xenon (~100kg)
- Oxygen free copper: (31cm)³
- 54 of low-BG 2-inch PMT
 - Photo coverage ~16%
- MgF₂ window
- 0.6 p.e. / keV

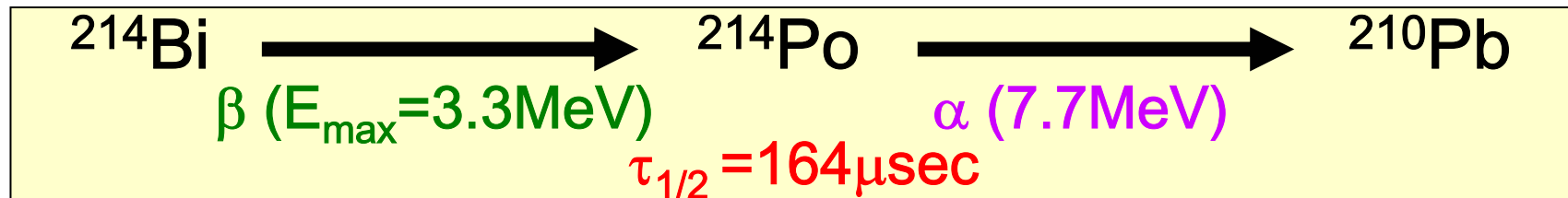


- Polyethylene (15cm)
- Boric acid (5cm)
- Lead (15cm)
- EVOH sheets (30mm)
- OFC (5cm)
- Rn free air (~3mBq/m³)

Rn assay with prototype detector

■ ^{238}U series

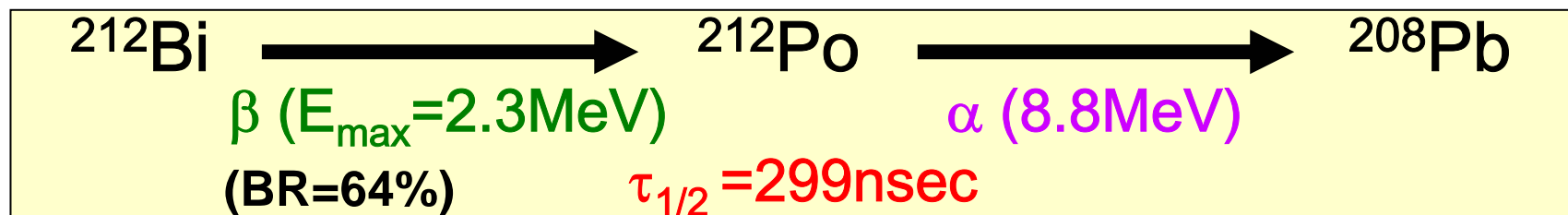
- ^{222}Rn ($\tau_{1/2} = 3.8\text{d}$, 3.3MeV beta), ...



➔ Observed coincident events

■ ^{232}Th series

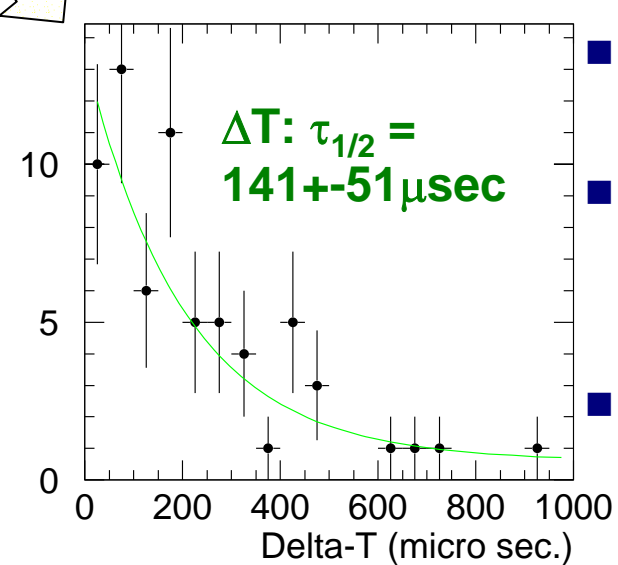
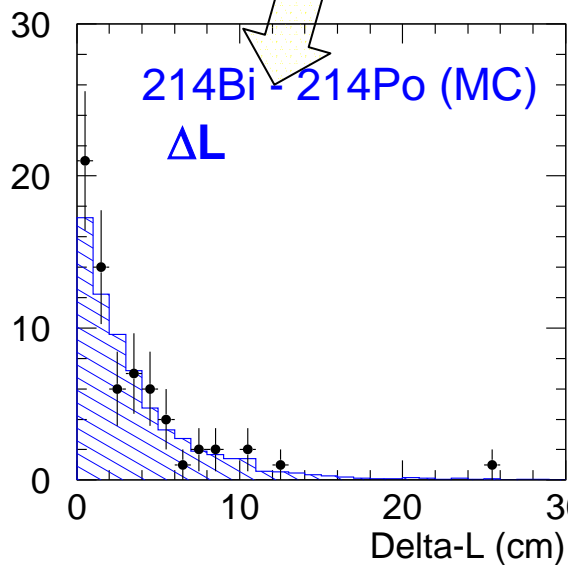
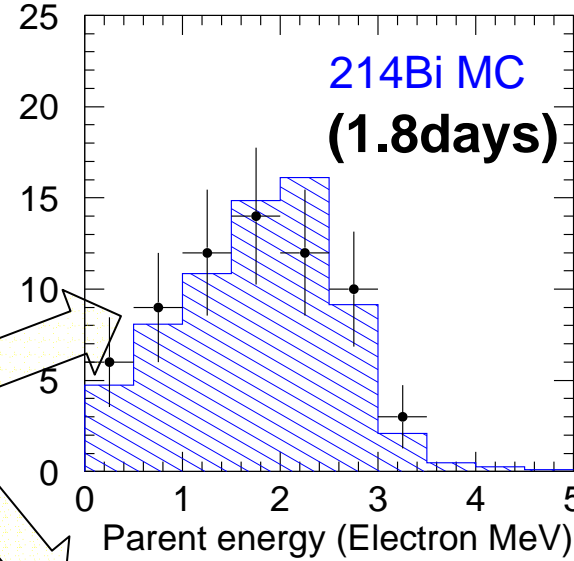
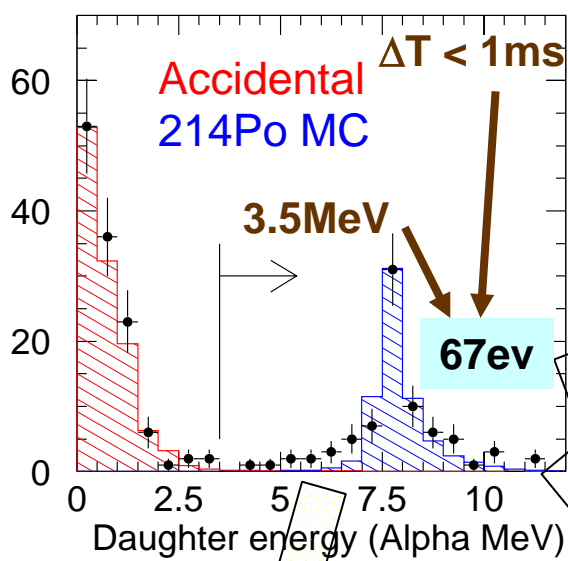
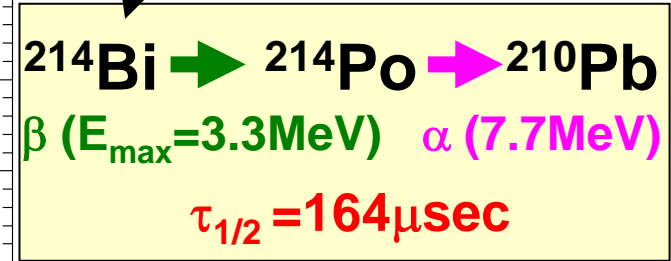
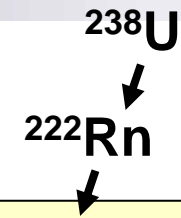
- ^{220}Rn (55s, 2.3MeV beta[64%]), ...



➔ No candidate found

^{222}Rn measurements

Aug.04 run
Preliminary



- 2 separate runs to check ^{222}Rn decay ($\tau_{1/2}=3.8\text{day}$)
- 4th Aug. 0.8day
 $^{238}\text{U}=(72\pm 11)\times 10^{-14}\text{ g/g}$
- 10th Aug. 1.0day
 $^{238}\text{U}=(33\pm 7)\times 10^{-14}\text{ g/g}$
- Consistent with expected ^{222}Rn decay ($(30\pm 5)\times 10^{-14}$)

Preliminary

Internal background sources

■ Current results

□ ^{238}U : = $(33 \pm 7) \times 10^{-14}$ g/g

Factor ~30, but may decay out further

□ ^{232}Th : < 23×10^{-14} g/g

Factor <~10, under further study

□ Kr : $\sim 3.3 \pm 1.1$ ppt

Would be achieved by an improved distillation system

Goal (~1ton)

1×10^{-14} g/g

2×10^{-14} g/g

1 ppt

Summary

- **A distillation system of Xe for Kr** was built in Kamioka Observatory. **~1/1000 Kr reduction / 1pass** was achieved.
 - **GC + API-MS system** was used for Kr assay in purified Xe.
 - ^{222}Rn and ^{220}Rn in purified Xe was measured by **XMASS prototype detector**.
 - The current **remaining impurities** in purified Xe for XMASS are below:
 - ^{238}U : = **$(33\pm 7)\times 10^{-14}$ g/g**
 - ^{232}Th : < **23×10^{-14} g/g**
 - **Kr**: = **3.3 ± 1.1 ppt**
- (assuming radiative equilibrium)