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 directory 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 N_2 can be used
- Various purification method
  - Distillation, circulation during experiment, …
  - Effective reduction against internal backgrounds
- No long life radioactive isotopes
- ^{136}Xe is a $\beta\beta$ decay candidate

Internal backgrounds

- **^{238}U series**
  - **^{222}Rn($\tau_{1/2}$ = 3.8d, 3.3MeV beta), …**
  - **Target (for XMASS 800kg detector): 1x10^{-14}g(^{238}U)/g(Xe)**
- **^{232}Th series**
  - **^{220}Rn(55s, 2.3MeV beta[64%]), …**
  - **Target: 2x10^{-14}g(^{232}Th)/g(Xe)**
- **^{85}Kr**
  - **Contamination during manufacture and refinement**
  - **$\tau_{1/2}$ = 10.8y, 687keV beta (99.6%)**
  - **Target: Kr = 1 ppt (mol)** (assuming ^{85}Kr/Kr = 1.2x10^{-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

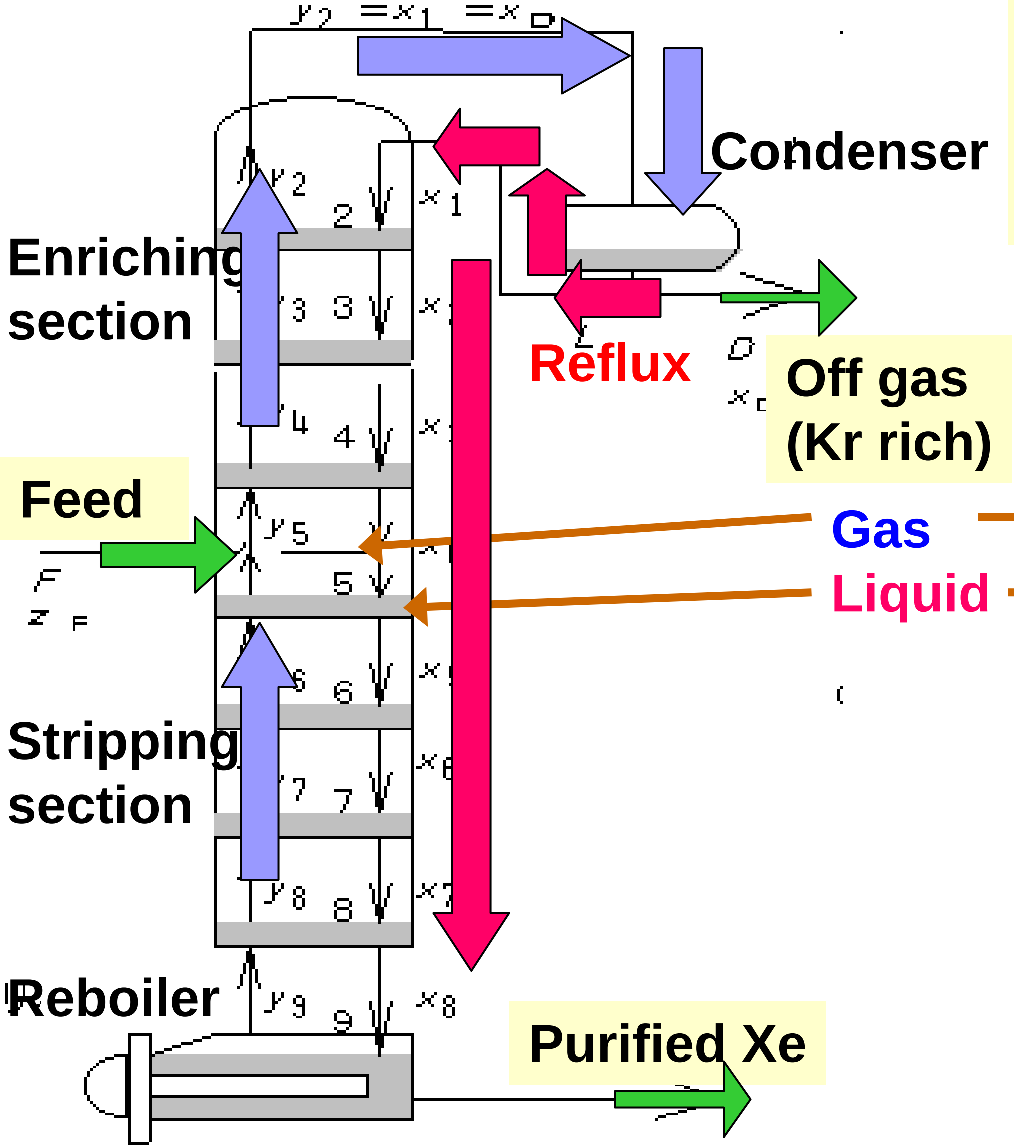
- Impurities in xenon
  - CO_2, H_2O : removed by adsorption
  - Kr, O_2, N_2, H_2, He : removed by distillation (boiling points are lower than Xe)

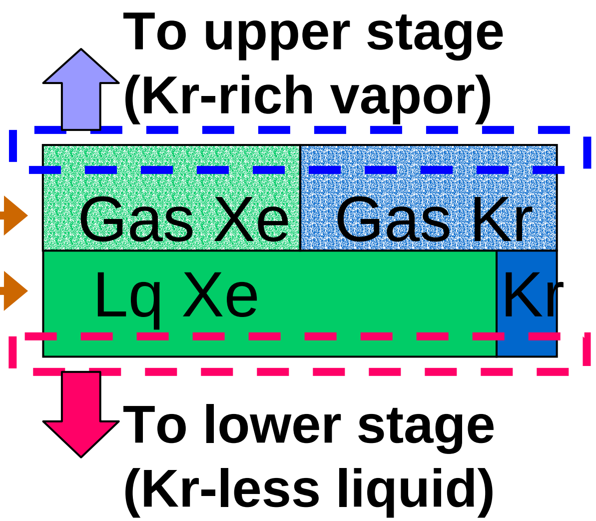
	Boiling point (@1 atm)	Boiling point (@2 atm)
Xe	165K	178K
Kr	120K	129K

- 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

= (Kr in gas / Kr in lq) /
(Xe in gas / Xe in lq)

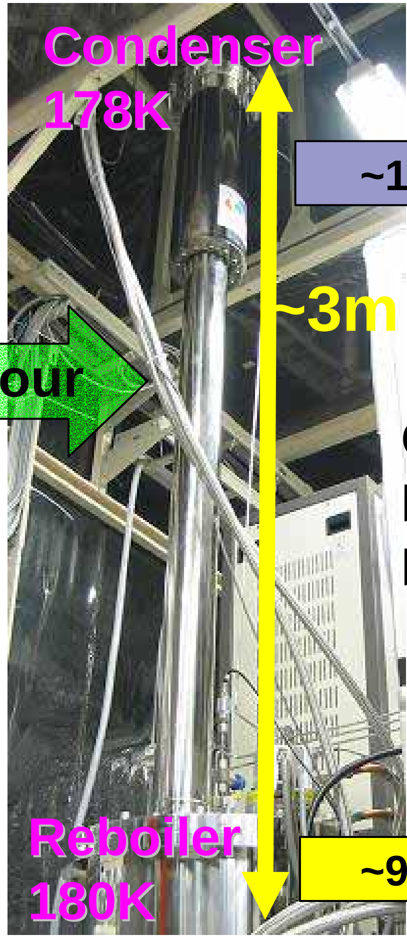
= 10.4 (@178K)

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

Raw Xe:
~3 ppb Kr

0.6 kg / hour



~1%

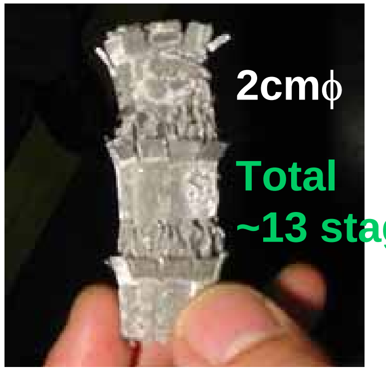
Off gas Xe:
330±100 ppb Kr
(measured)

Operation: 2 atm
Reflux ratio = ~190
Design factor: 1/1000 Kr / 1 pass

~99%

Purified Xe:
3.3±1.1 ppt Kr
(measured)

Sulzer Chemtech:
EX laboratory packing



Kr assay with API-MS detector

GC + API-MS system

Measurement of Kr in purified Xe

Kr assay result

GC + API-MS system

This system was developed at Taiyo Toyo Sanso Co. Ltd. (SAAN). http://www.saan.co.jp/english/

- API: Atmospheric Pressure Ionization
- 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

$$C^+ + X \rightarrow C + X^+$$

(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

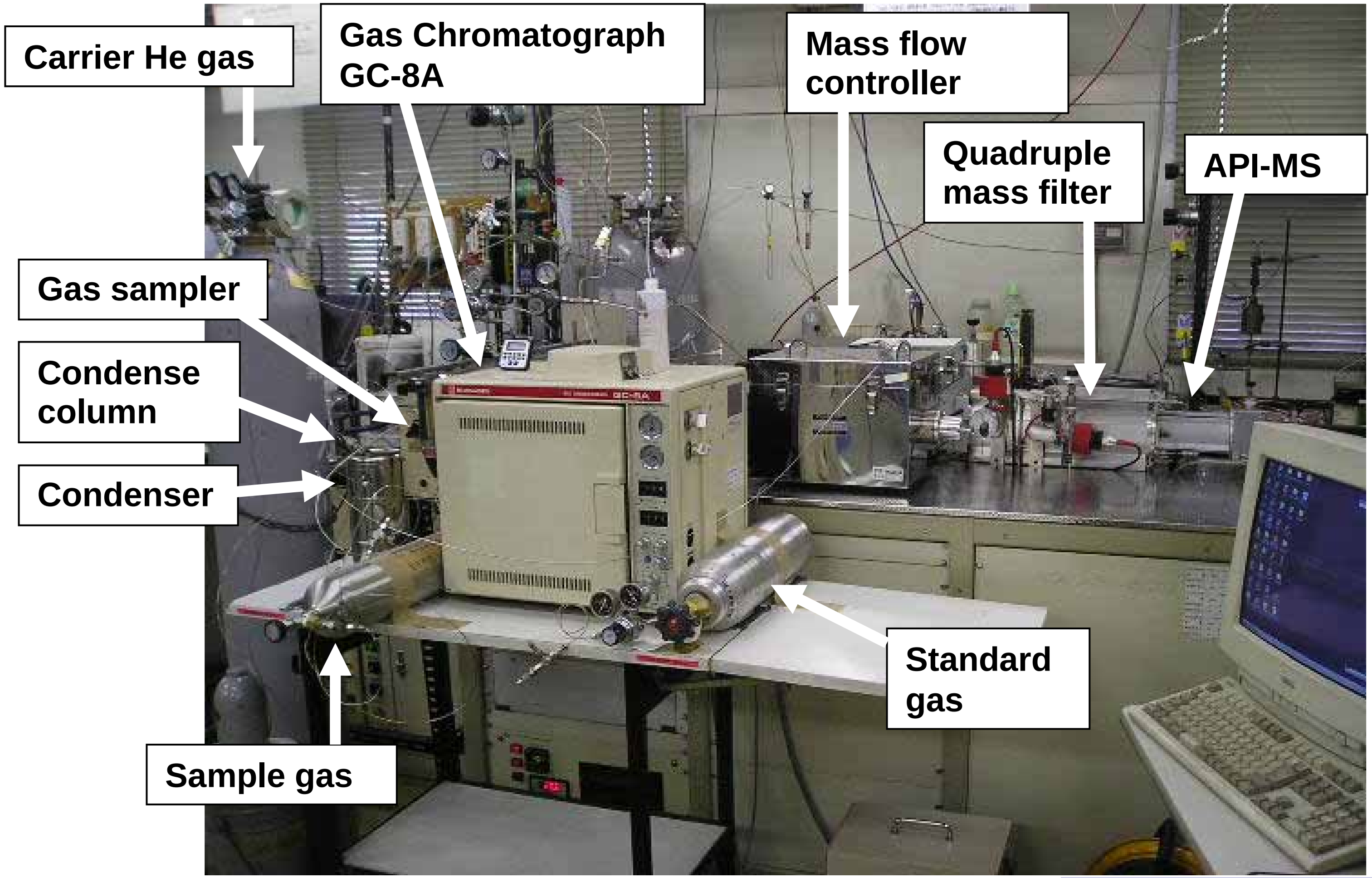
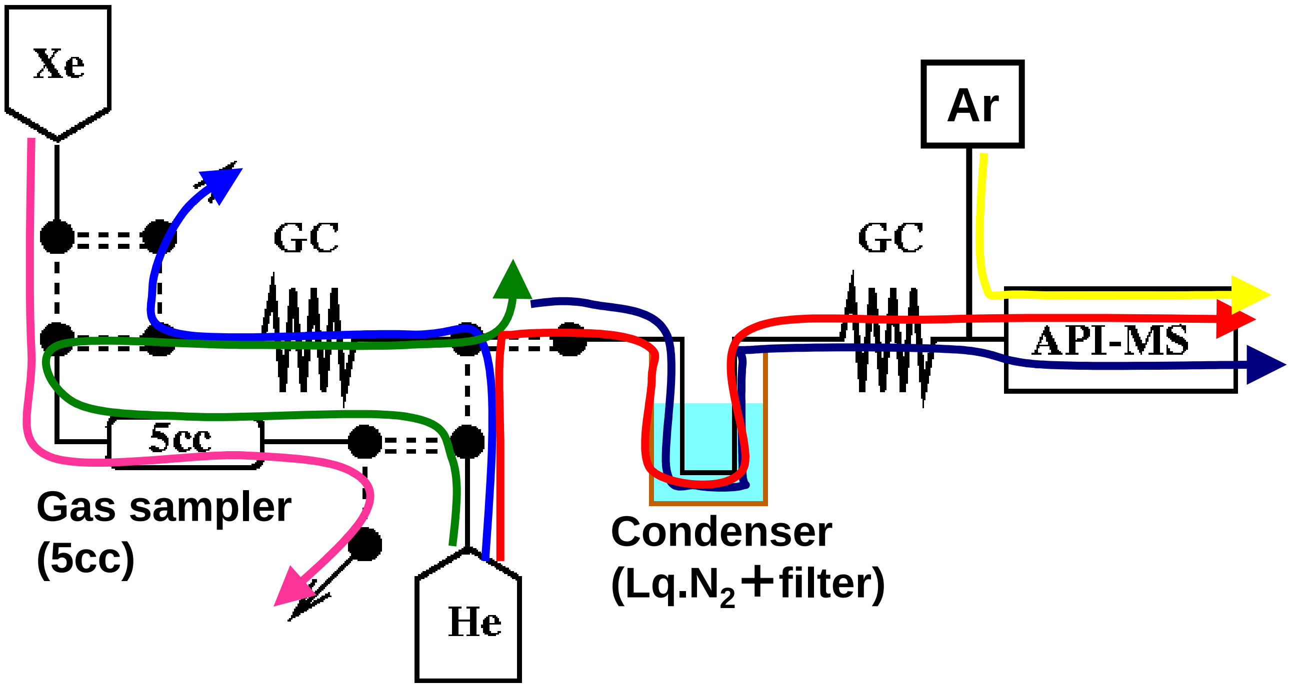


Photo by Kawaguchi Laboratory, Research & Development Division, Taiyo Toyo Sanso Co. Ltd.

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



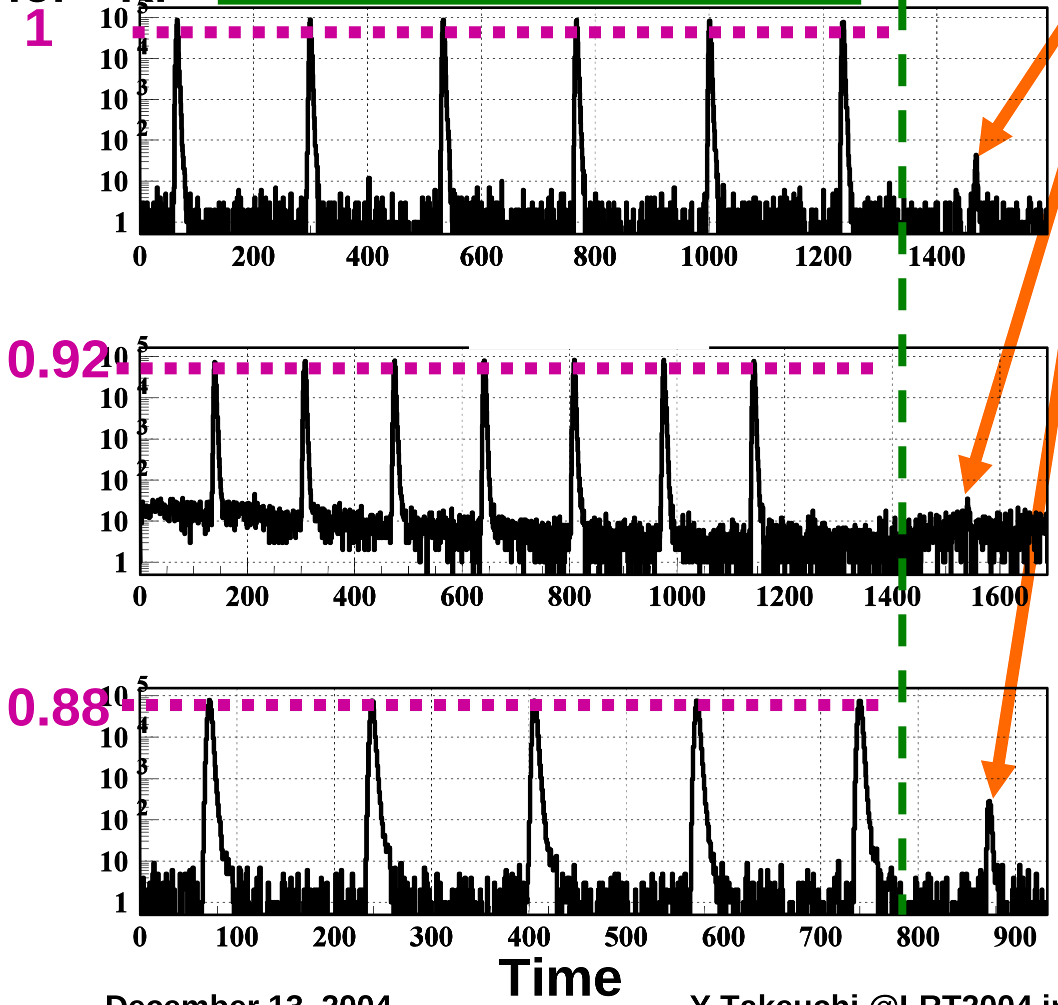
- Purge GC with He
- Sample 5cc Xe
- Feed sample Xe into GC with He carrier
- Only Kr timing, feed sample gas into the condenser.
- Repeat above
- After heat up, feed condensed Kr into API-MS (select only Kr timing by GC)

Kr assay result

Measurement after 100 times condense

API-MS count for ^{84}Kr

Calibration with 1ppm Kr standard gas



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

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

Factor ~1/1000 / 1pass was achieved

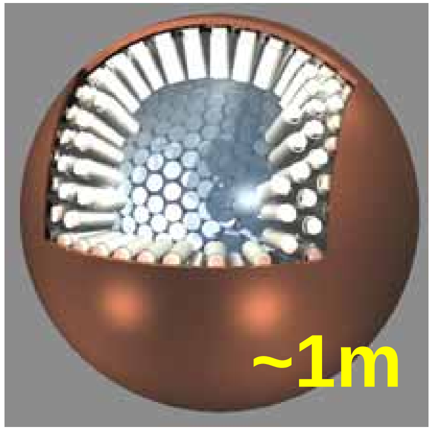
Rn assay with XMASS prototype detector

Strategy of the XMASS project
XMASS prototype detector
Rn assay with prototype detector
^{222}Rn measurements
Internal background sources

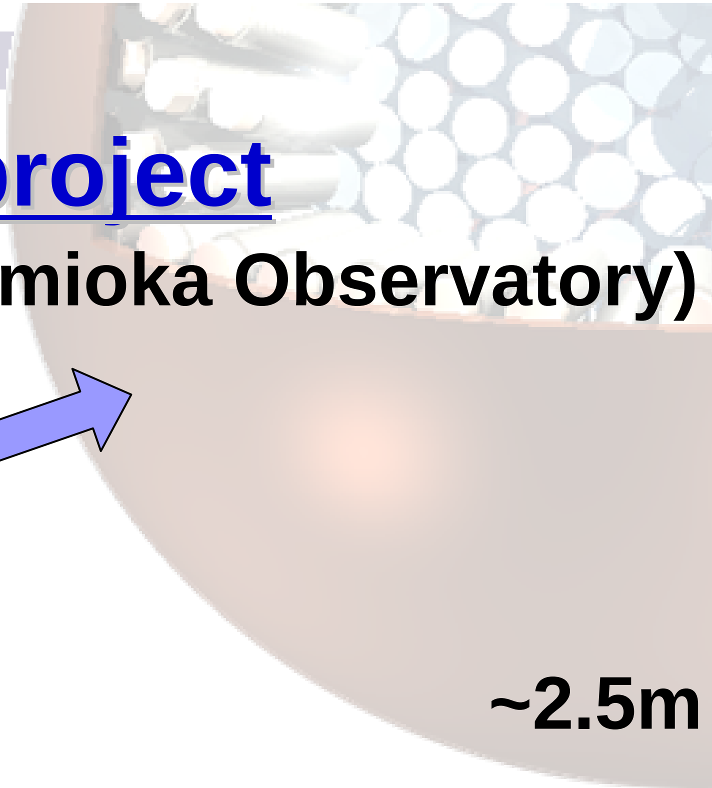
Strategy of the XMASS project

(in Kamioka Observatory)

Prototype detector
(FV 3kg) R&D

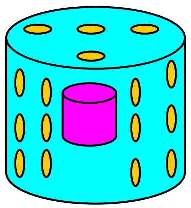


~1 ton detector
(FV 100kg)
Dark matter search



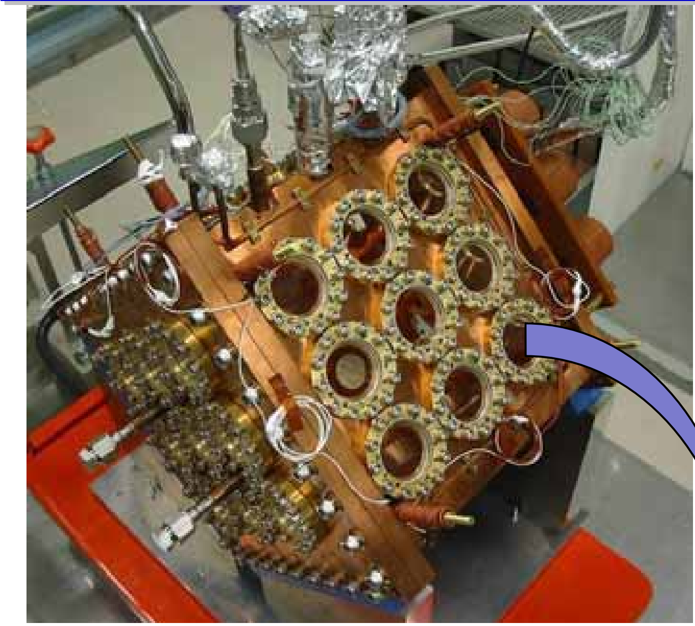
~20 ton detector
(FV 10ton)
Solar neutrinos
Dark matter search

Confirmation of feasibilities of the ~1 ton detector
- Analysis techniques
- Self shielding performance
- Low background properties
- Purification techniques

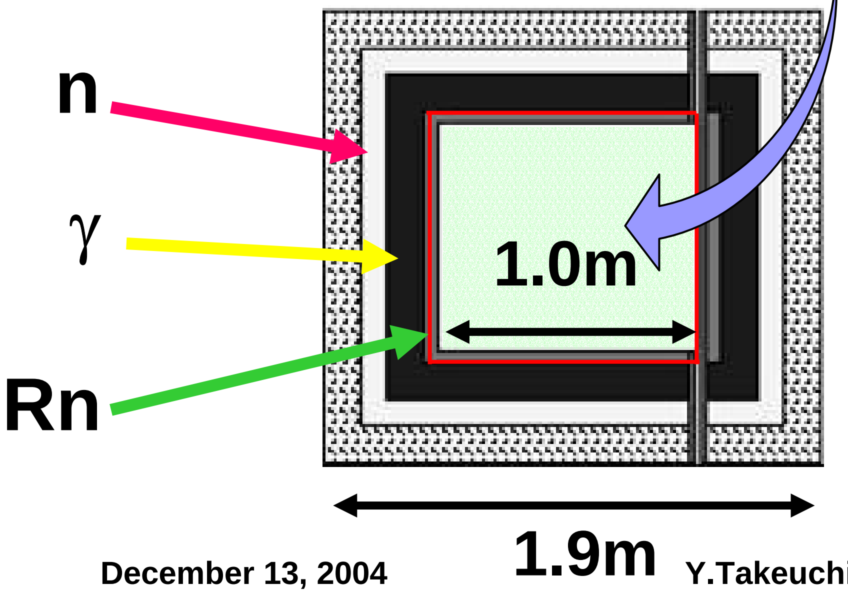


Dedicated detector for
Double beta decay search

XMASS prototype detector



- 30 litter liquid Xenon (~100kg)
- Oxygen free copper: (31cm)^3
- 54 of low-BG 2-inch PMT
  - Photo coverage ~16%
- MgF_2 window
- 0.6 p.e. / keV



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

Rn assay with prototype detector

- **^{238}U series**
  - **^{222}Rn($\tau_{1/2}$ = 3.8d, 3.3MeV beta), …**

^{214}Bi → ^{214}Po → ^{210}Pb

β (E_{max}=3.3MeV) α (7.7MeV)

$\tau_{1/2}$ =164μsec

⇨ Observed coincident events

- **^{232}Th series**
  - **^{220}Rn(55s, 2.3MeV beta[64%]), …**

^{212}Bi → ^{212}Po → ^{208}Pb

β (E_{max}=2.3MeV) α (8.8MeV)

(BR=64%)

$\tau_{1/2}$ =299nsec

⇨ No candidate found

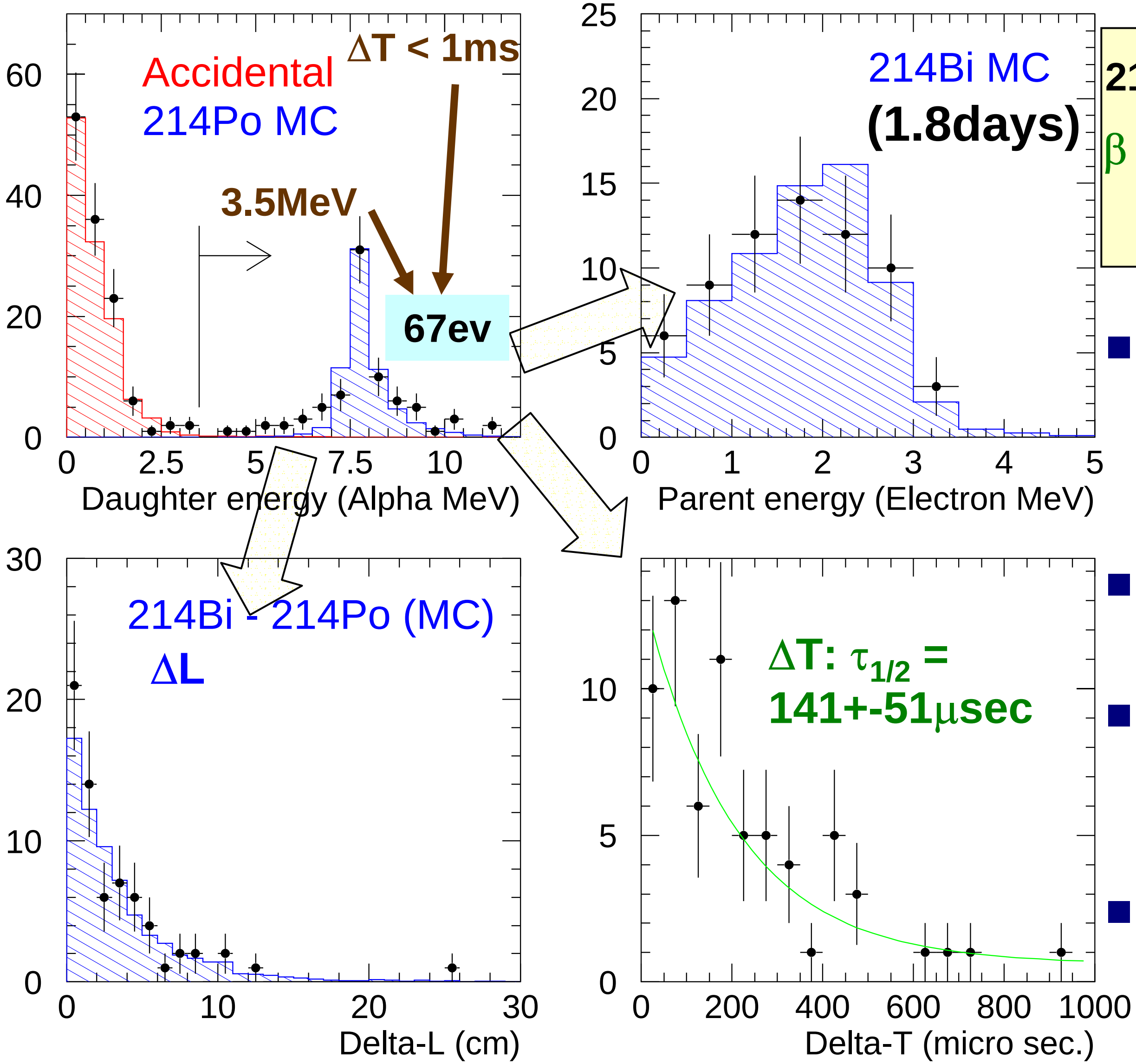
^{222}Rn measurements

Aug.04 run
Preliminary

^{238}U → ^{222}Rn → ^{214}Bi → ^{214}Po → ^{210}Pb

β (E_{max}=3.3MeV) α (7.7MeV)

$\tau_{1/2}$ =164μsec



- 2 separate runs to check ^{222}Rn decay ($\tau_{1/2}$=3.8day)
- 4th Aug. 0.8day
  ^{238}U=(72+-11)x10^{-14} g/g
- 10th Aug. 1.0day
  ^{238}U=(33+-7)x10^{-14} g/g
  (assuming radiative equilibrium)
- Consistent with expected ^{222}Rn decay ((30+-5)x10^{-14})

Internal background sources

Preliminary

- **Current results**
  - ^{238}U: = (33+-7)x10^{-14} g/g

    •Factor ~30, but may decay out further
    •Planning to install a Rn-dedicated purification device

  - ^{232}Th: < 23x10^{-14} g/g

    Factor <~10, under further study

  - Kr: ~3.3±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 ppt level 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 the prototype detector are below:
  - ^{238}U: = $(33+-7)\times10^{-14}$ g/g
  - ^{232}Th: $<$ 23×10^{-14} g/g
  - Kr: = 3.3 ± 1.1 ppt

(assuming radiative equilibrium)