Rn assay and distillation purification of Lq. Xe for Kr

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

- 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 N₂ can be used
- Various purification method
  - Distillation, circulation during experiment, ...
  - Effective reduction against internal backgrounds
- No long life radioactive isotopes
  - \(^{136}\text{Xe}\) is a \(\beta\beta\) decay candidate

<table>
<thead>
<tr>
<th>Scintillation light</th>
<th>~42photon/keV</th>
</tr>
</thead>
<tbody>
<tr>
<td>Scintillation light wave length</td>
<td>175nm</td>
</tr>
<tr>
<td>Scintillation light width</td>
<td>~40nsec</td>
</tr>
<tr>
<td>Atomic number</td>
<td>54</td>
</tr>
<tr>
<td>Atomic weight</td>
<td>131.29 amu</td>
</tr>
<tr>
<td>Density</td>
<td>3.0 g/cm³</td>
</tr>
<tr>
<td>Melting (boiling) point</td>
<td>161.4K (165.1K)</td>
</tr>
<tr>
<td>Chemical series</td>
<td>Noble gases</td>
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</tbody>
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Noble gases

161.4K (165.1K) Melting (boiling) point

3.0 g/cm³ Density

131.29 amu Atomic weight

~40nsec Scintillation light width

175nm Scintillation light wave length

54 Atomic number

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Internal backgrounds

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

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

- **85Kr**
  - Contamination during manufacture and refinement
  - $\tau_{1/2} = 10.8\text{y}, 687\text{keV beta}(99.6\%)$
  - 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)

<table>
<thead>
<tr>
<th></th>
<th>Boiling point (@1 atm)</th>
<th>Boiling point (@2 atm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Xe</td>
<td>165K</td>
<td>178K</td>
</tr>
<tr>
<td>Kr</td>
<td>120K</td>
<td>129K</td>
</tr>
</tbody>
</table>

- 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

\[
\text{Relative volatility} = \frac{(\text{Kr in gas} / \text{Kr in lq})}{(\text{Xe in gas} / \text{Xe in lq})} = 10.4 \ (@178K)
\]
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

Sulzer Chemtech: EX laboratory packing

2cm\(\phi\)
Total ~13 stages

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

Condenser 178K

~3m

~1% to ~99%

Reboiler 180K

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

Purified Xe: 3.3 ± 1.1 ppt Kr (measured)

0.6 kg / hour

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Kr assay with API-MS detector

GC + API-MS system
Measurement of Kr in purified Xe
Kr assay result
GC + API-MS system

- **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
  
  \[
  C^+ + X \rightarrow C + X^+
  \]

- **Ionization potential**:
<table>
<thead>
<tr>
<th></th>
<th>He</th>
<th>Ar</th>
<th>Kr</th>
<th>Xe</th>
</tr>
</thead>
<tbody>
<tr>
<td>eV</td>
<td>24.6</td>
<td>15.8</td>
<td>14.0</td>
<td>12.3</td>
</tr>
</tbody>
</table>

- **Need to extract Kr from Xe**

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

Carrier He gas
Gas Chromatograph GC-8A
Mass flow controller
Quadruple mass filter
API-MS
Gas sampler
Condense column
Condenser
Sample gas
Standard gas

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

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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**

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 = 
\[
\frac{(\text{Sample Xe gas} - \text{He gas})}{\text{Kr 50ppt gas} \times 50\text{ppt}}
\]

= \(3.3 ± 1.1\) ppt (stat. error only)

Factor ~1/1000 / 1pass was achieved

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Rn assay with XMASS prototype detector

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

(in Kamioka Observatory)

Dedicated detector for Double beta decay search

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

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

NOW

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

Prototype detector
(FV 3kg) R&D

~30cm

~1m

~2.5m

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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$^3$)

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Rn assay with prototype detector

**238U series**

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

  - $^{214}\text{Bi}$ → $^{214}\text{Po}$ → $^{210}\text{Pb}$
    - $\beta$ ($E_{\text{max}} = 3.3\text{MeV}$)
    - $\alpha$ ($7.7\text{MeV}$)
    - $\tau_{1/2} = 164\mu\text{sec}$

  - Observed coincident events

**232Th series**

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

  - $^{212}\text{Bi}$ → $^{212}\text{Po}$ → $^{208}\text{Pb}$
    - $\beta$ ($E_{\text{max}} = 2.3\text{MeV}$)
    - $\alpha$ ($8.8\text{MeV}$)
    - $\tau_{1/2} = 299\text{nsec}$

  - No candidate found
222Rn measurements

214Bi → 214Po → 210Pb
β (E_{max}=3.3MeV)  α (7.7MeV)
τ_{1/2} =164µsec

2 separate runs to check 222Rn decay
(τ_{1/2}=3.8day)

4th Aug. 0.8day
238U=(72+-11)x10^{-14} g/g

10th Aug. 1.0day
238U=(33+-7)x10^{-14} g/g
(assuming radiative equilibrium)

Consistent with expected 222Rn decay
((30+-5)x10^{-14} )

214Bi 214Po (MC)
ΔL

ΔT < 1ms
3.5MeV
67ev

ΔT: τ_{1/2} = 141+-51µsec

214Bi MC
(1.8days)

238U
222Rn

Accidental
214Po MC

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Internal background sources

Current results

- $^{238}\text{U} : \ket\approx (33\pm 7) \times 10^{-14} \text{ g/g}$
  - Factor $\sim 30$, but may decay out further
  - Planning to install a Rn-dedicated purification device

- $^{232}\text{Th} : < 23 \times 10^{-14} \text{ g/g}$
  - Factor $<\sim 10$, under further study

- Kr: $\sim 3.3 \pm 1.1 \text{ ppt}$
  - Would be achieved by an improved distillation system

Goal (~1 ton)
- $1 \times 10^{-14} \text{ g/g}$
- $2 \times 10^{-14} \text{ g/g}$
- 1 ppt
Summary

- A distillation system of Xe for Kr was built in Kamioka Observatory. ~1/1000 Kr reduction / 1 pass 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:  = $(33\pm 7) \times 10^{-14}$ g/g (assuming radiative equilibrium)
  - $^{232}$Th:  < $23 \times 10^{-14}$ g/g
  - Kr:  = $3.3 \pm 1.1$ ppt