Purification of Scintillating Crystals

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CANDLES Collaboration

Outline

Purification of Materials

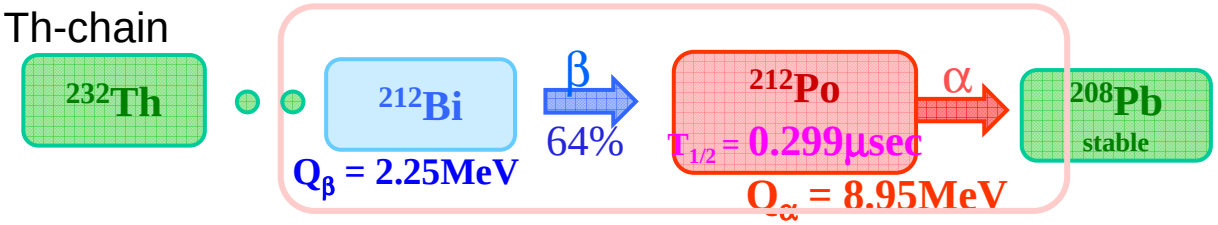
- Events from Contaminations within CaF_2(pure) Scintillators
  - In CANDLES system . . .
- Studies for High Purity Crystals
  - Selection of CaF_2 Powder
  - Rinse of CaF_2 Powder
- Next Step
- Summary

Effects from Contamination in CaF_2 Scintillators

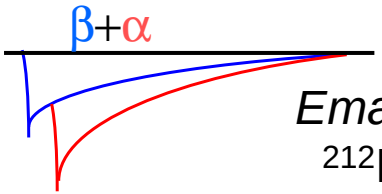
Serious Backgrounds

Pile-up Events from Sequential Decays

Th-chain



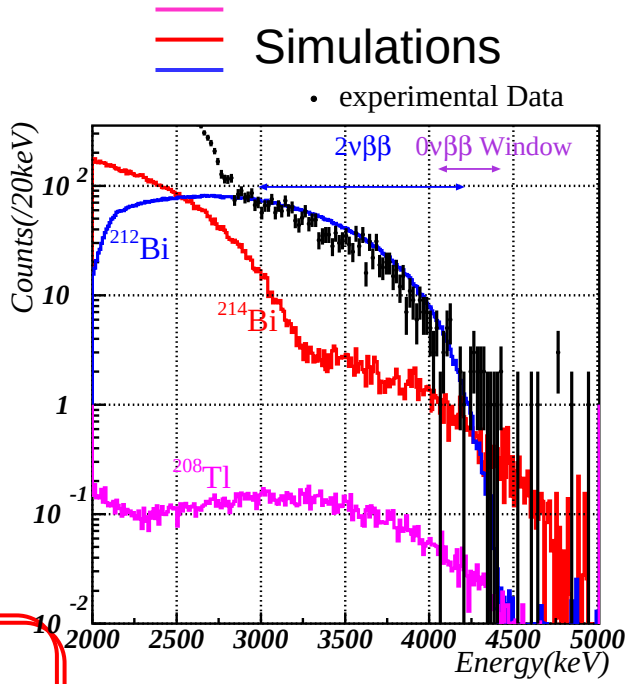
Deposit Energy from Pile-up Events



Emax=5.3MeV
$^{212}Bi \rightarrow ^{212}Po \rightarrow ^{208}Pb$(Th-chain)

Emax=5.8MeV
$^{214}Bi \rightarrow ^{214}Po \rightarrow ^{210}Pb$(U-chain)

Background Study
for ELEGANT VI system



Effects from Contaminations

- Main Backgrounds around Q-value Region

⇩

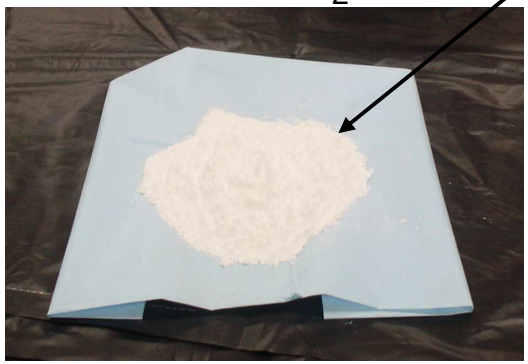
- Reduction → Background Free Measurement

Development of High Purity Crystals

Growing Process of CaF_2(pure) Crystals

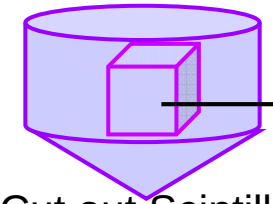
Raw Materials $CaCO_3$、HF etc → CaF_2 Powder → Sintering CaF_2 Melting CaF_2 → CaF_2 Crystals

1）Selection of CaF_2 Powder
2）Rinse of CaF_2 Powder

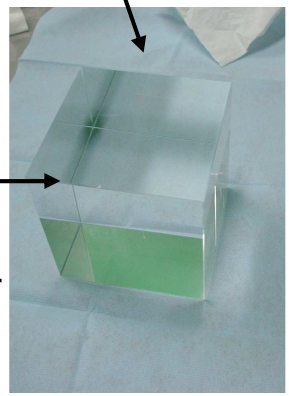


(in Solid State)

Cut out
Polish



Cut out Scintillator from CaF_2 Ingot



For High Purity Crystals…

- Selection of CaF_2 Powder
  - Ge Detector
- Rinse of CaF_2 Powder

↔ Comparison with Radioactivities in Crystals

Measurement of Radioactivities in CaF_2 Crystals

Delayed Coincidence Measurement

at Oto Cosmo Observatory

U-chain

^{238}U ($T_{1/2}=4.5\times10^{9}$year) → ^{214}Bi (Q_β =3.27MeV) —β→ ^{214}Po ($T_{1/2}$=164μsec) —α→ (Q_α = 7.83MeV) → ^{210}Pb → ^{206}Pb stable

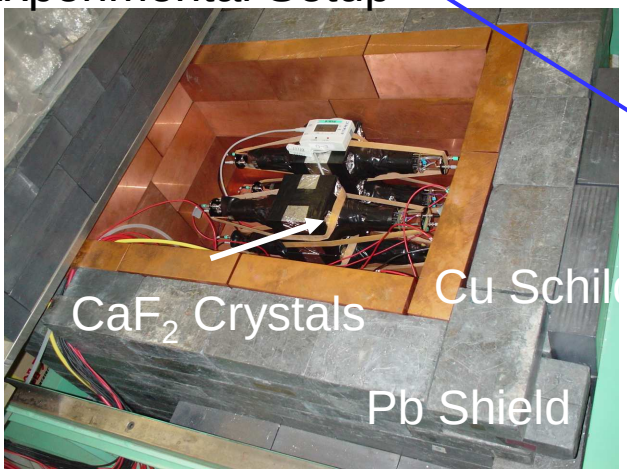
Th-chain

^{232}Th ($T_{1/2}=1.4\times10^{10}$year) → ^{220}Rn (Q_α = 6.28MeV) —α→ ^{216}Po ($T_{1/2}$=145msec) —α→ (Q_α = 6.78MeV) → ^{212}Pb → ^{208}Pb stable

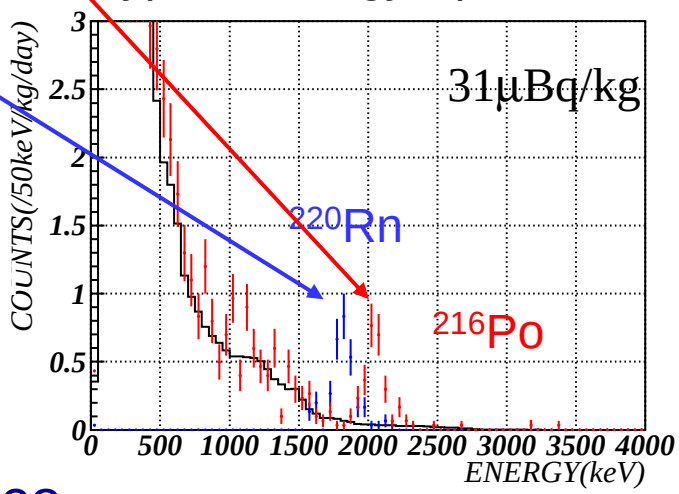
Detection Efficiencies

- U-chain : ～90%
- Th-chain : ～85%

Experimental Setup



Typical Energy Spectra



Delayed Coincidence …

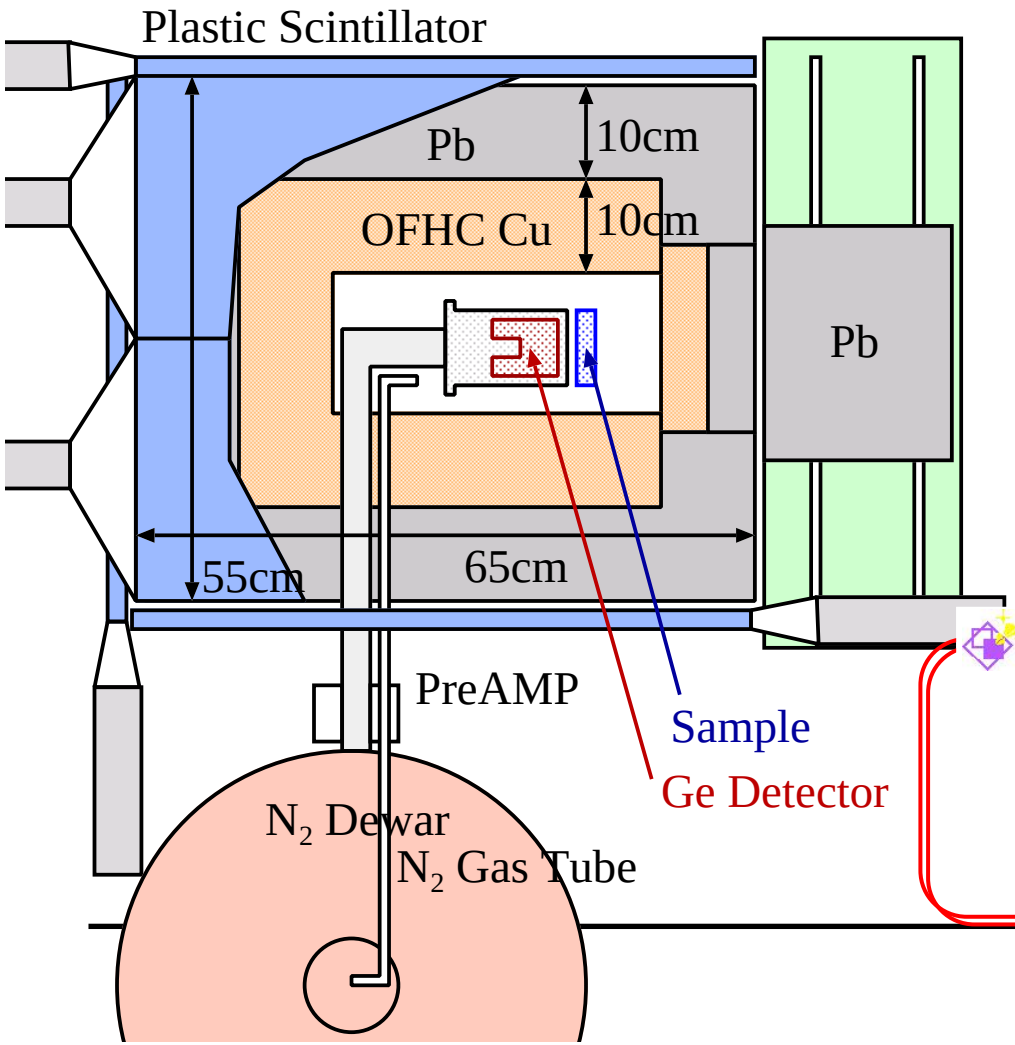
- Sensitivity : ～5μBq/kg
  Req. for CANDLES III ＝100μBq/kg

Measurement of Radioactivities in CaF_2 Powder

Ge Semiconductor Detector

at Osaka Univ.
（Sea Level Laboratory）

ELEGANT III System (Ge Detector)



Ge Detector : 171cc

Shields
Cu 10cm 、Pb 10cm

Plastic Scintillator (6 Modules)
as Veto Counters

Air Tight Film for Rn Purge
Filled by N_2 gas

Sensitivities …

^{226}Rn(U-chain)	:	～3mBq/kg
Th-chain	:	～3mBq/kg
^{40}K	:	～15mBq/kg

Selection of CaF_2 Powder

Collaboration with
OHYO KOKEN CO.,LTD.

Selection of CaF_2 Powder

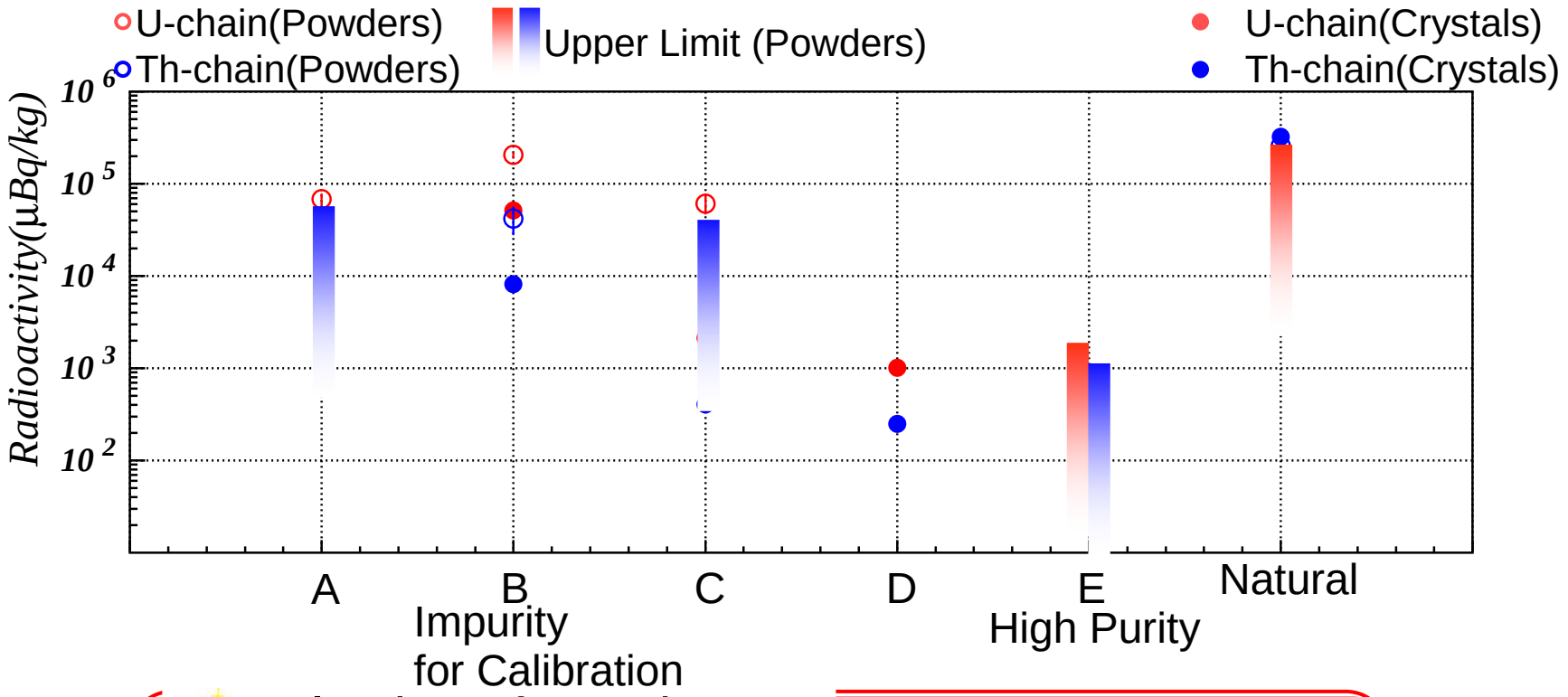
- Measurement Radioactivities of CaF_2 Powder and CaF_2 Crystals
  - Crystals from B、C、D、E and Natural Powders …5 Crystals
    Delayed Coincidence Measurement (α-rays counting)
    - Crystal Size … B: $2.8 \times 2.8 \times 1.8 cm^3$
      C: $5 \times 5 \times 5 cm^3$
      D: $3 \times 3 \times 3 cm^3$
      E: $5 \times 5 \times 2 cm^3$
      Natural: $3.1\phi \times 1.1 cm^3$
  - Powder A、B、C、E、Natural Powder…5 Powders
    Measurement with Ge Detector (γ-rays counting)

Selection of CaF_2 Powder

Selection of CaF_2 Powder

Collaboration with
OHYO KOKEN CO.,LTD.

Radioactivities of CaF_2 Powders and Crystals



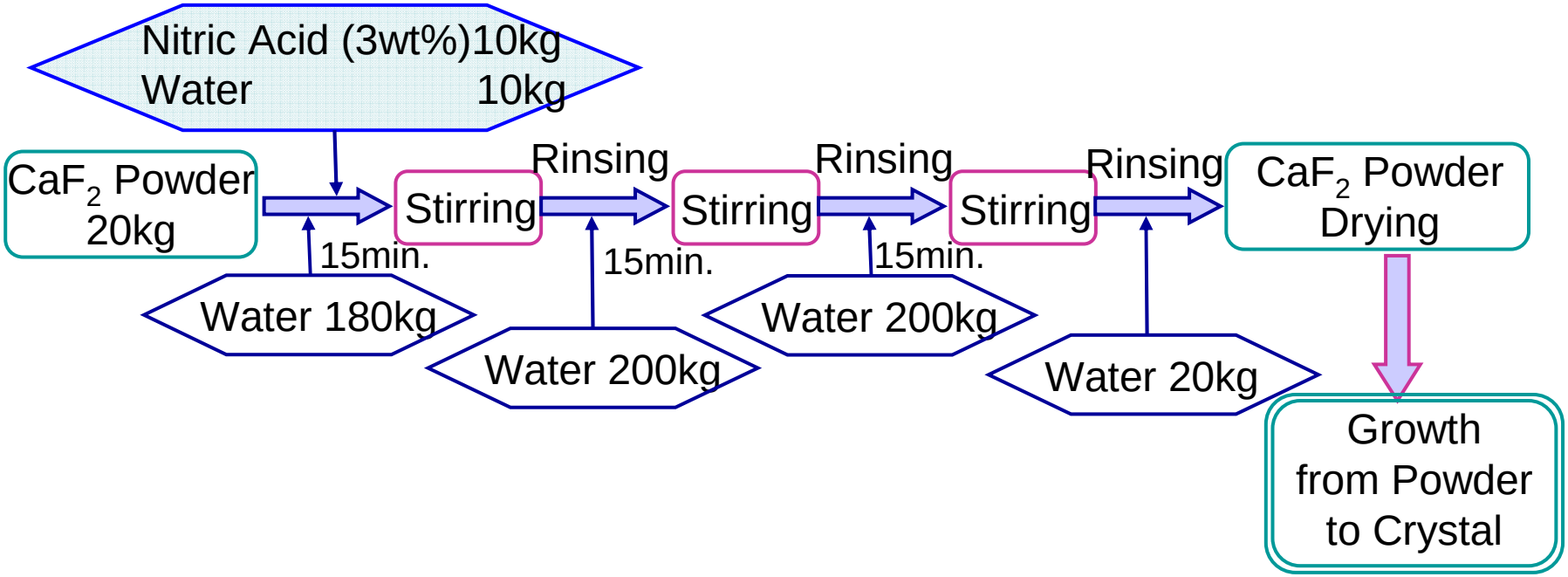
- Selection of Powders …
  - Very Effective（100mBq/kg～10μBq/kg）
  - High Purity or Impurity CaF_2 by Powder Selection

Rinse of CaF_2 Powder

Rinse of CaF_2 Powder

Collaboration with
OHYO KOKEN CO.,LTD.

- Rinse . . .
  - CaF_2 Powder 20kg : for 1 CaF_2 Crystal of 10cm Cube
  - Rinse with Nitric Acid



Rinse of CaF_2 Powder

Rinse of CaF_2 Powder

Collaboration with
OHYO KOKEN CO.,LTD.

Reduction of Contaminations

Before Rinse

^{214}Po(U-chain) : $1.12 \pm 0.03(stat.)^{+0.10}_{-0.12}(syst.)$ mBq/kg
^{212}Po(Th-chain) : 1.67 ± 0.04 mBq/kg
^{215}Po(Ac-chain) : $1.69 \pm 0.03(stat.)^{+0.30}_{-0.35}(syst.)$ mBq/kg

After Rinse

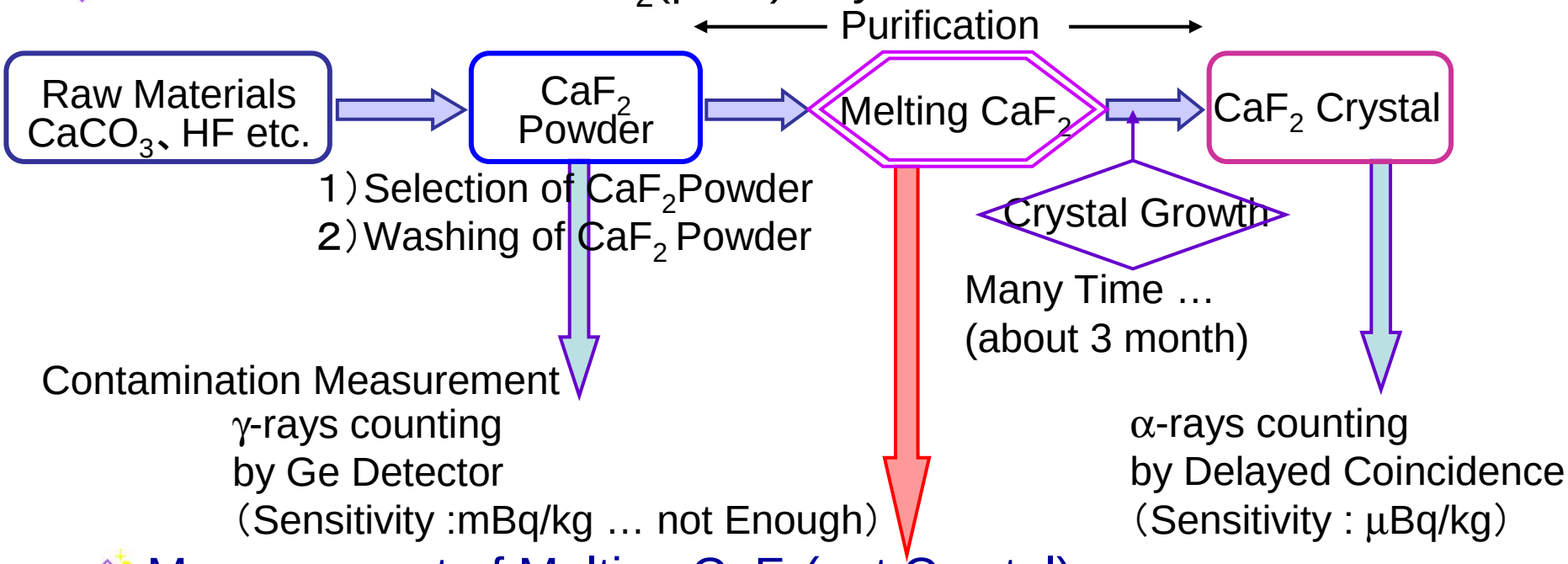
^{214}Po(U-chain) : $0.07 \pm 0.02(stat.)^{+0.01}_{-0.02}(syst.)$ mBq/kg
^{212}Po(Th-chain) : 0.95 ± 0.03 mBq/kg
^{215}Po(Ac-chain) : $0.70 \pm 0.03(stat.)^{+0.13}_{-0.14}(syst.)$ mBq/kg

- Effect of Waching . . .
  - Washing with Nitric Acid
  - Effective for U-chain Contamination
  - Why only U-chain ?? → Next Step

Next Step for High Purity Crystals

Contamination in Melting CaF_2

- Growth Process of CaF_2(pure) Crystals



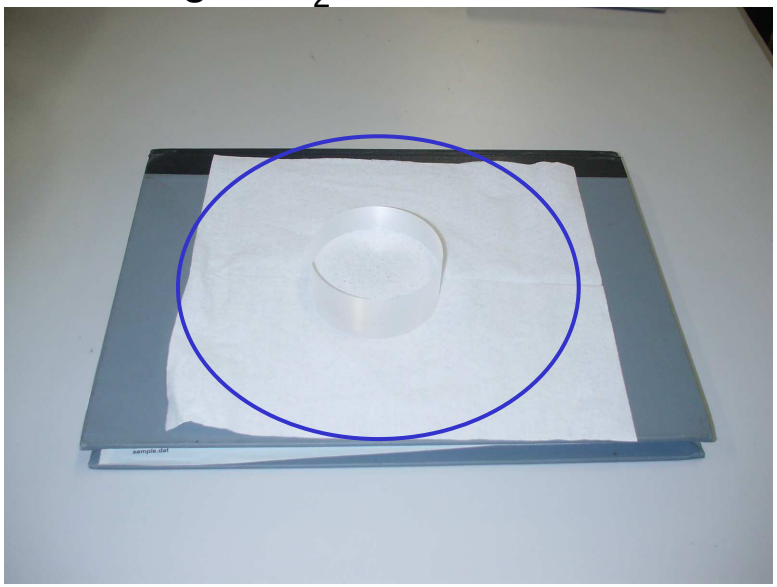
- Measurement of Melting CaF_2(not Crystal)
  - Possibility of α-ray Measurement
    - ⇩ if Melting CaF_2 Scintillates
  - Good Sensitivity than γ-rays Counting

Measurement of Melting CaF_2

Sample of Melted CaF_2

Collaboration with
Stella Chemifa Co.

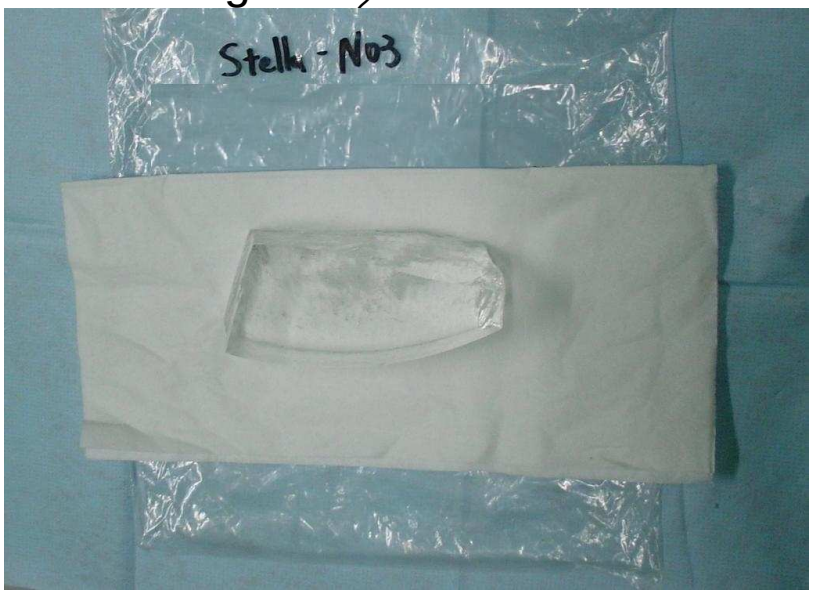
Melting CaF_2 1



Stella-N02(367.5g)
(7ϕ × 3cm³)

Sample of High Purity Melting CaF_2
(by ICP-MS Measurement)

Melting CaF_2 2



Stella-N03（341.7g）

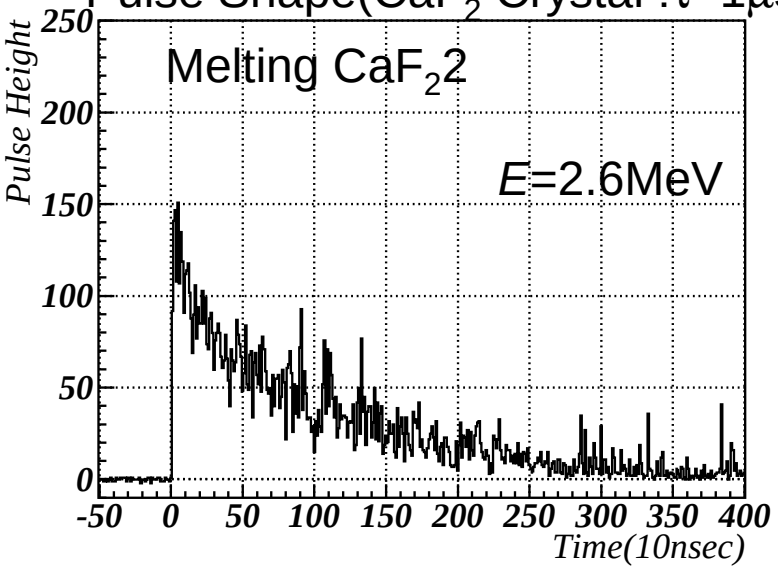
Sample of Impurity Melting CaF_2
(by ICP-MS Measurement)

Measurement of Melting CaF_2

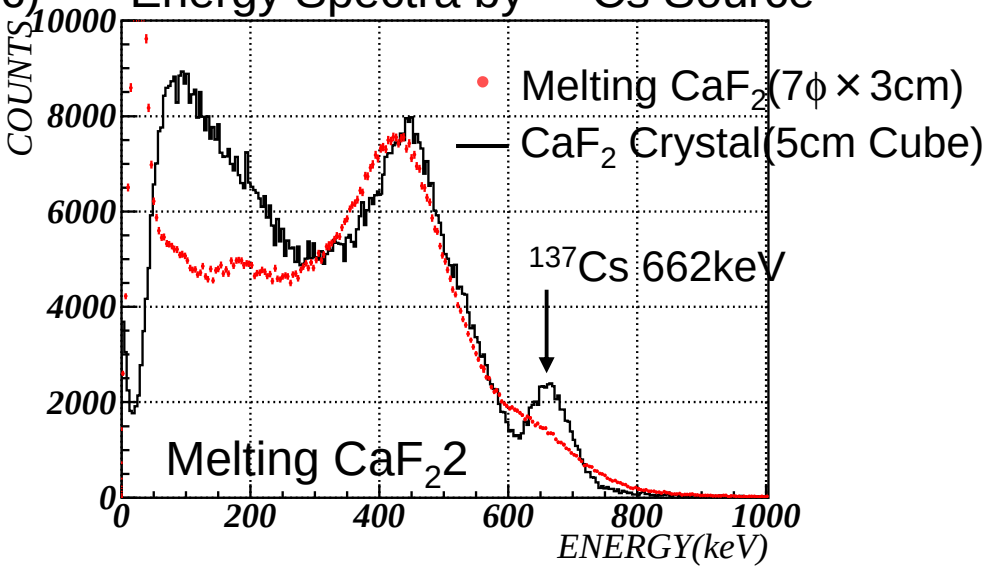
Collaboration with
Stella Chemifa Co.

Scintillation

Pulse Shape(CaF_2 Crystal :τ=1μsec)



Energy Spectra by ^{137}Cs Source



Measurement of Melting CaF_2

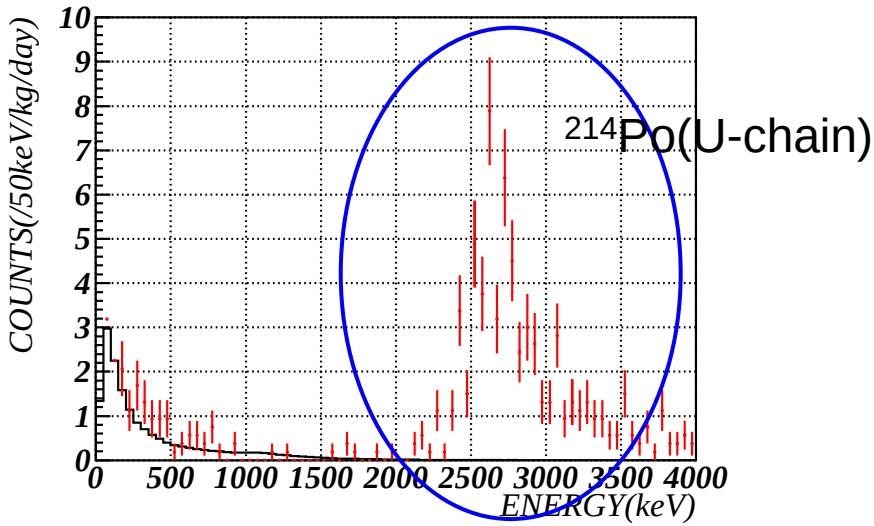
- Scintillate
- Worse Energy Resolution
  due to Worse Light Collection

Measurement of Melting CaF_2

Measurement of Radioactive Contaminations

Collaboration with
Stella Chemifa Co.

Melting CaF_2 1 : Stella-N02(7ϕ × 3cm^3 368g)



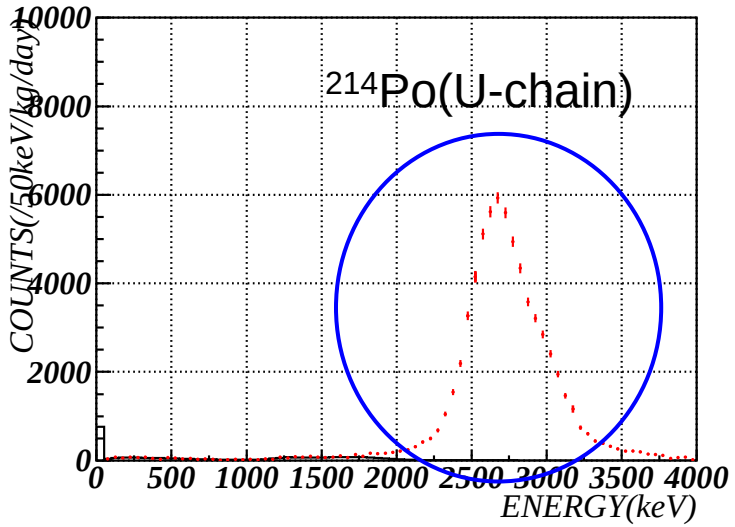
If in Radiative Equilibrium

^{214}Po(U-chain) : 0.87±0.05mBq/kg
(0.071±0.004ppb)

^{216}Po(Th-chain) : 0.083±0.020mBq/kg
(0.020±0.005ppb)

Measuring Time : 14.5 days

Melting CaF_2 2 : Stella-N03(342g)



If in Radiative Equilibrium

^{214}Po(U-chain) : 927±6mBq/kg
(75.0±0.5ppb)

^{216}Po(Th-chain) : <9.1mBq/kg
(<2.2ppb)

Measuring Time : 1.0 days

Measurement of Melting CaF_2

Collaboration with
Stella Chemifa Co.

Comparison with ICP-MS Results

α-rays Measurement
（Assumption of Radiative Equilibrium）

Melting CaF_2 2

^{214}Po(U-chain) : 75.0±0.5 ppb
^{212}Po(Th-chain) : <2.2 ppb

Melting CaF_2 3

^{214}Po(U-chain) : 100.1±0.7 ppb
^{212}Po(Th-chain) : <3.3 ppb

⟺ Consistent

ICP-MS

U : 80 ppb
Th : <20 ppb

U : 100 ppb
Th : <20 ppb

Sensitivities for High Purity CaF_2
U ～0.001ppb
Th ～0.002ppb

Sensitivities for High Purity CaF_2
U 0.2ppb
Th 0.3ppb

Measurement of Melting CaF_2

- Enable to Measure Contaminations
- Consistent with ICP-MS Results
- To Next Step for High Purity Crystals

Summary

- Contaminations within CaF_2 Crystals
- Study of High Purity Crystals
  - Selection of CaF_2 Powder
  - Rinse of CaF_2 Powder
- To Next Step
  - Measurement of Melting CaF_2