

# Low radioactivity $\text{CaF}_2$ scintillator crystals for CANDLES

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for the CANDLES collaboration

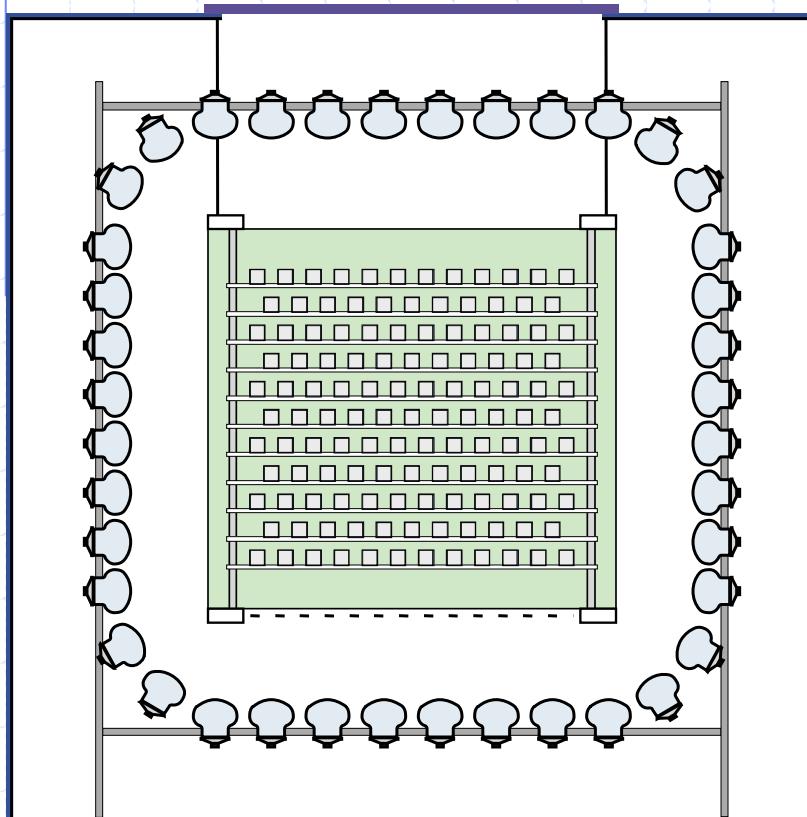
# Candles

# Double beta decay of $^{48}\text{Ca}$

- ◆ Largest Q value (4.27 MeV)
  - next largest;  $^{150}\text{Nd}$  (3.3 MeV)
  - large phase space factor
  - almost background free ( $\gamma$ : 2.6 MeV,  $\beta$ : 3.3 MeV)
- ◆ Low Natural abundance → 0.187%
  - large detector
  - enrichment
- ◆ Next generation detector : fight against BG!
  - $\langle m_\nu \rangle \propto T^{-1/2} \propto M_{\text{det}}^{-1/2}$  if background free
  - $\langle m_\nu \rangle \propto T^{-1/2} \propto M_{\text{det}}^{-1/4}$  if background limited

# CANDLES

CAlcium fluoride for studies of Neutrino and Dark matters  
by Low Energy Spectrometer



- ◆ undoped  $\text{CaF}_2$  ( $\text{CaF}_2(\text{pure})$ )
  - ${}^{48}\text{Ca}$  ( $Q_{\beta\beta} = 4.27 \text{ MeV}$ )
  - Atten. length > 1 m
  - Low radioactive impurities
- ◆ Low background detector
  - $4\pi$  active shield (LS)
  - Passive shield (Water, LS)
  - Pulse shape information
- ◆ Good energy resolution
  - large photo-coverage
  - Two phase LS system



Candles

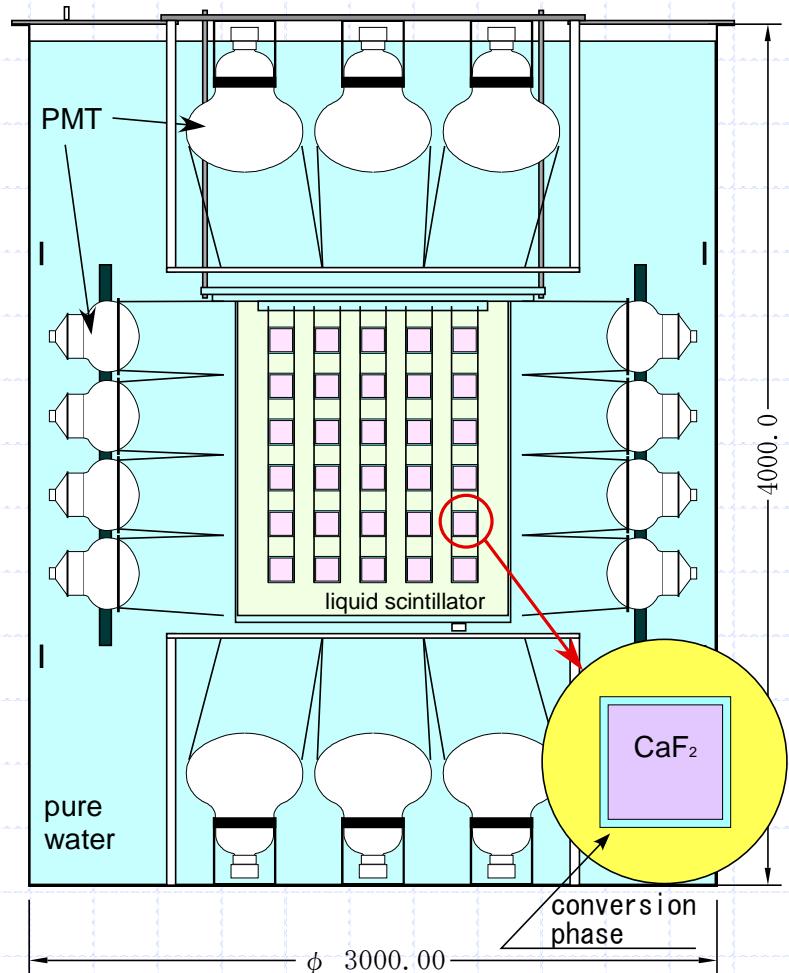
# CANDLES III (U.G.) @Kamioka



Candles

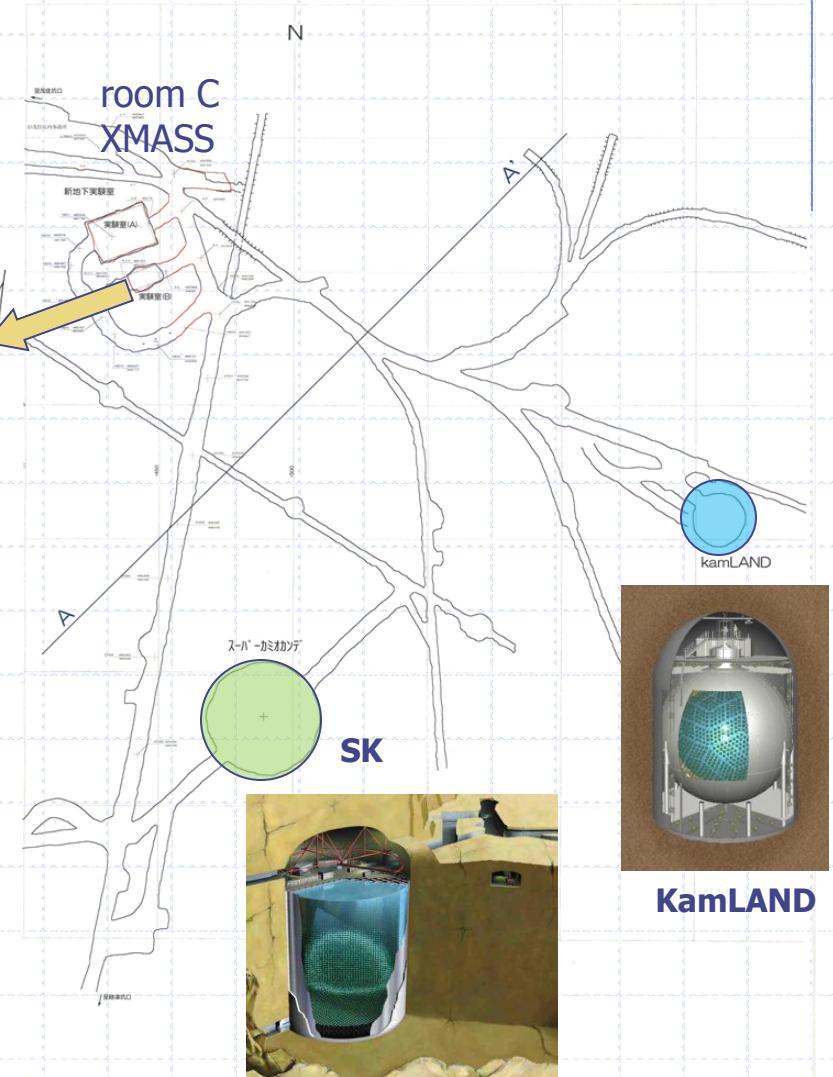
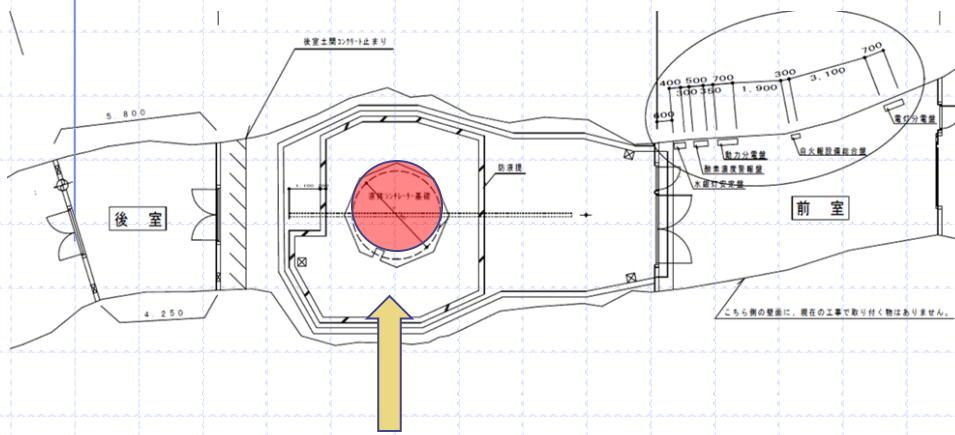
# CANDLES III(U.G.)

- ◆ CaF<sub>2</sub>(pure)
  - 10<sup>3</sup> cm<sup>3</sup> × 96 crystals; 305 kg
- ◆ Liquid scintillator
  - two phase system
  - Purification system
- ◆ H<sub>2</sub>O Buffer
  - passive shield (larger tank)
- ◆ PMTs
  - 17" PMT (× 14) : R7250
  - 13" PMT (× 56) : R8055
- ◆ photon trans. simulation
  - ➡ energy res. ~4.0 % @  $Q_{\beta\beta}$
- ◆ Kamioka underground lab.



# Kamioka new exp. room

## experimental room D



# CANDLES III (U.G.)



Candles





Candles

# Undoped CaF<sub>2</sub> scintillation crystal

# Energy resolution

- ◆ Keep high transparency for both( $\text{CaF}_2(\text{UV})$ ,  $\text{LS}(\text{vis.})$ ) scintillation light

$\text{CaF}_2$  crystal, LS, pure water, acrylic vessel,...

- Undoped  $\text{CaF}_2$  (attenuation length > 1m)
  - ◆ cf.  $\text{CaF}_2(\text{Eu}) \sim 10$  cm
- Shift wavelength of scintillation light from  $\text{CaF}_2$  scintillators; UV  $\Rightarrow$  visible

- ◆ Large photo-coverage

- Large (13,17 inch) PMT



Candles

# Two Phase System

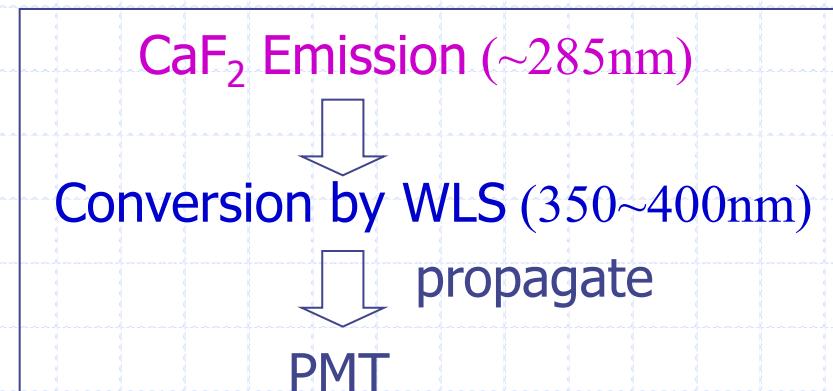
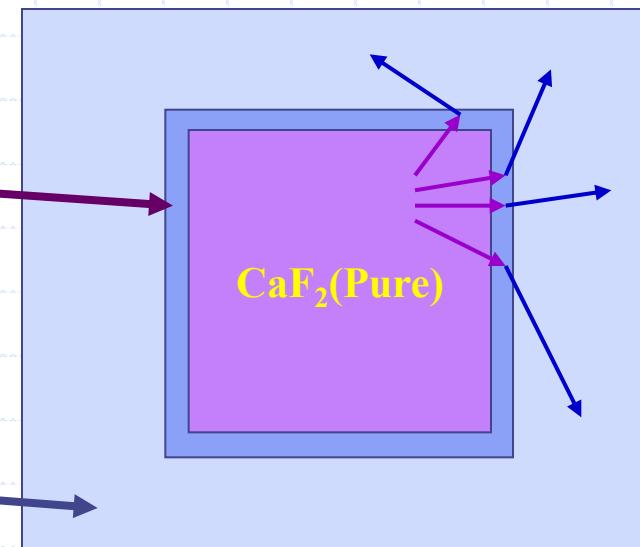
## ◆ Concept of Method

### ■ Conversion Phase

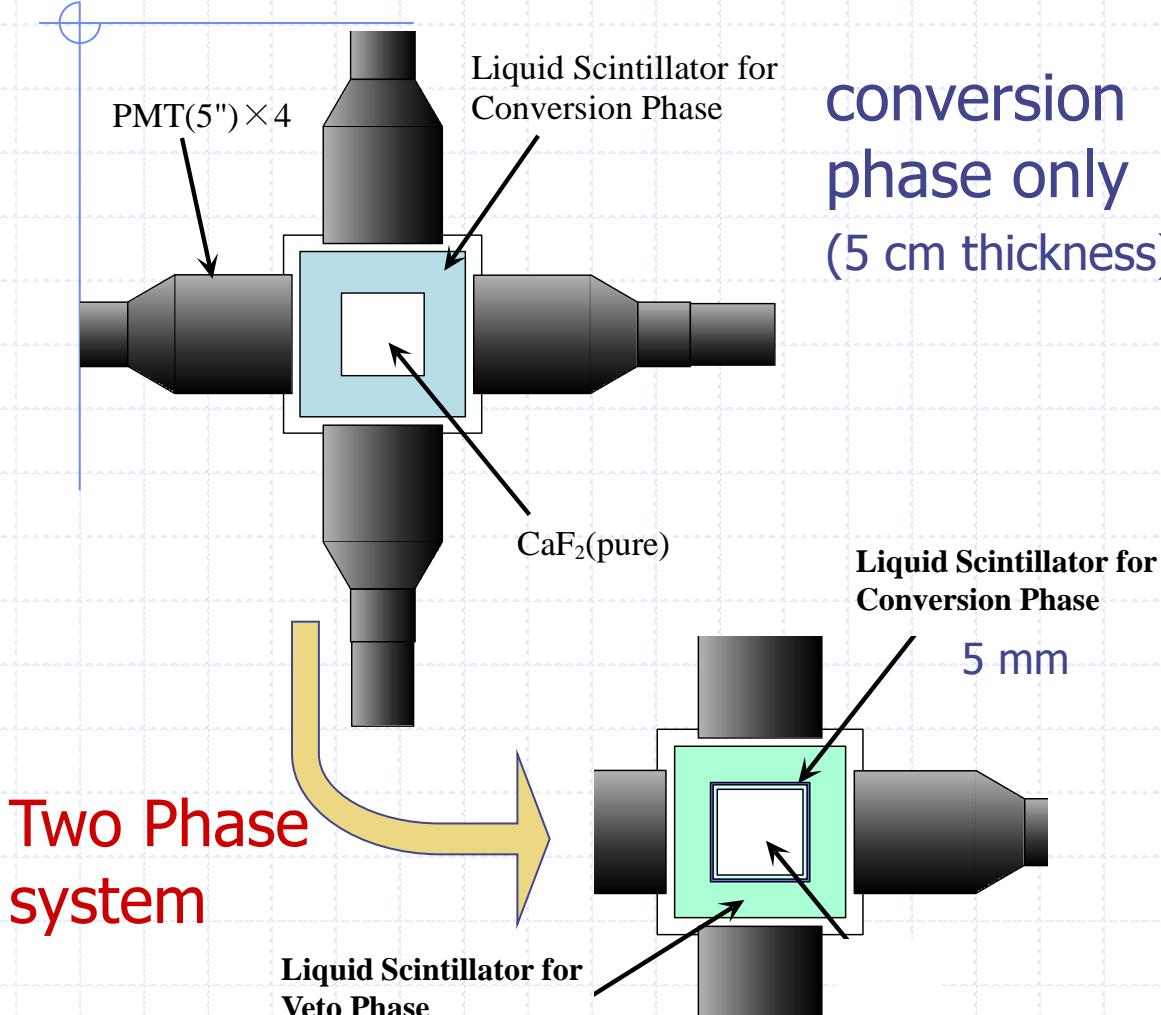
- ◆ large conversion eff.
- ◆ good transparency for UV

### ■ Veto Phase

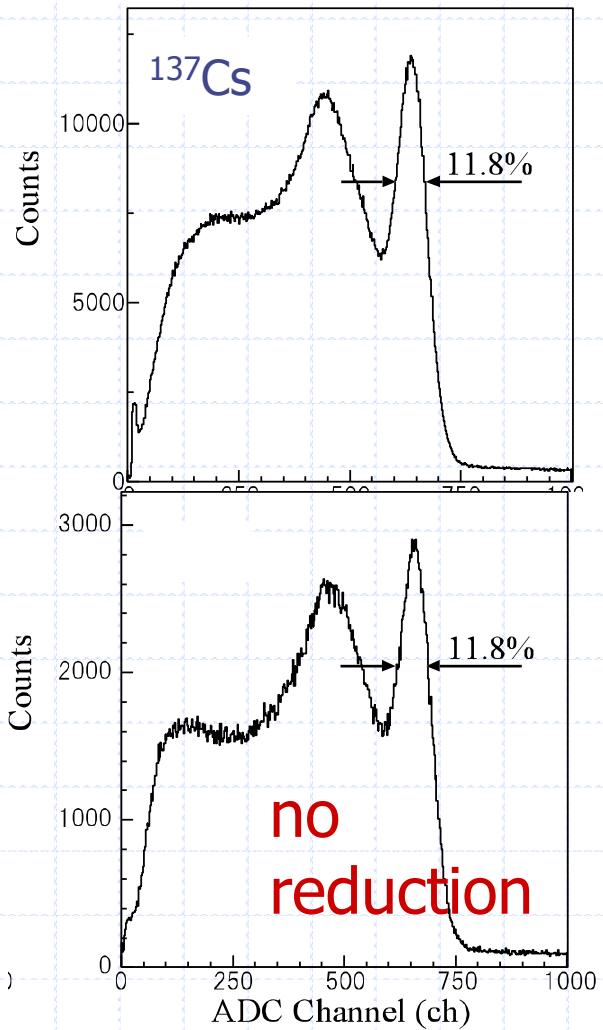
- ◆ large light output with aromatic solvent  
(absorb UV light)
- ◆ good transparency for visible light



# Performance of two phase system



conversion  
phase only  
(5 cm thickness)



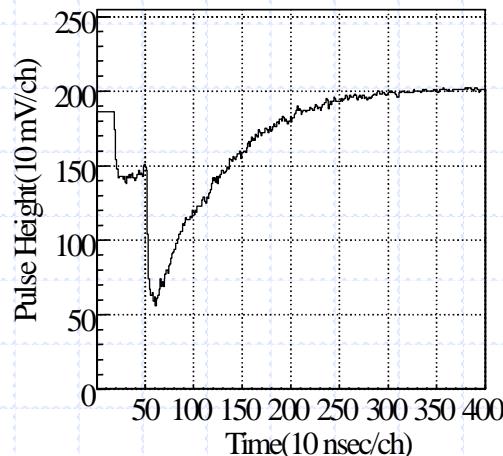


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# Main BG in CaF<sub>2</sub> scintillator

## ◆ BG

### ■ Successive decays in U, Th



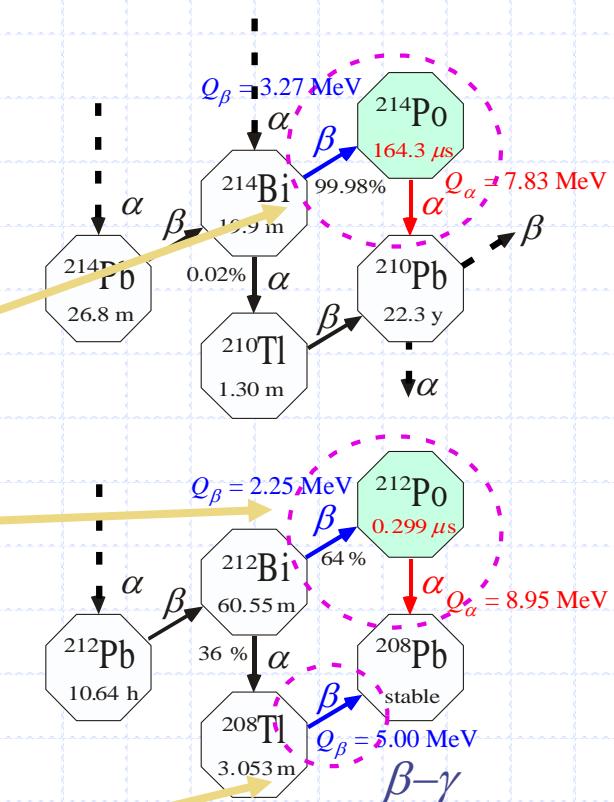
$$E_{vis} = Q_\beta + Q_\alpha \times f \approx Q_{\beta\beta}$$

f : Quenching factor for  $\alpha$

### ■ $^{208}\text{TI}$ : $\beta - \gamma$

$^{214}\text{Bi}$  (U)

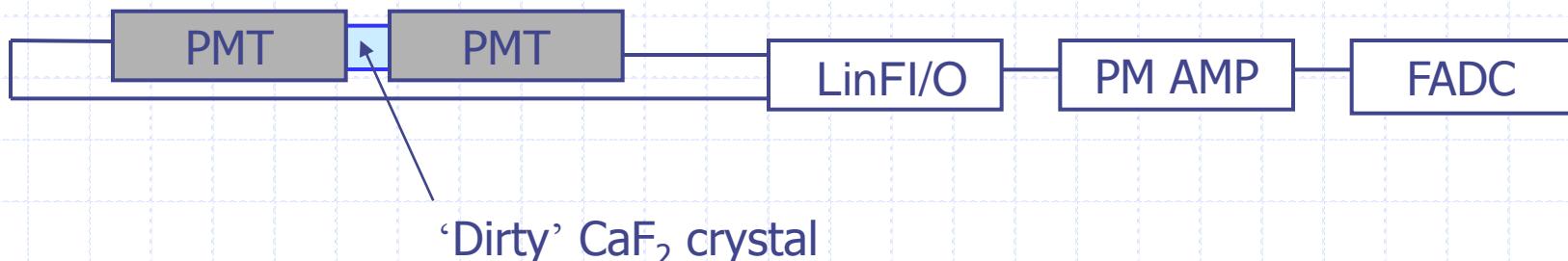
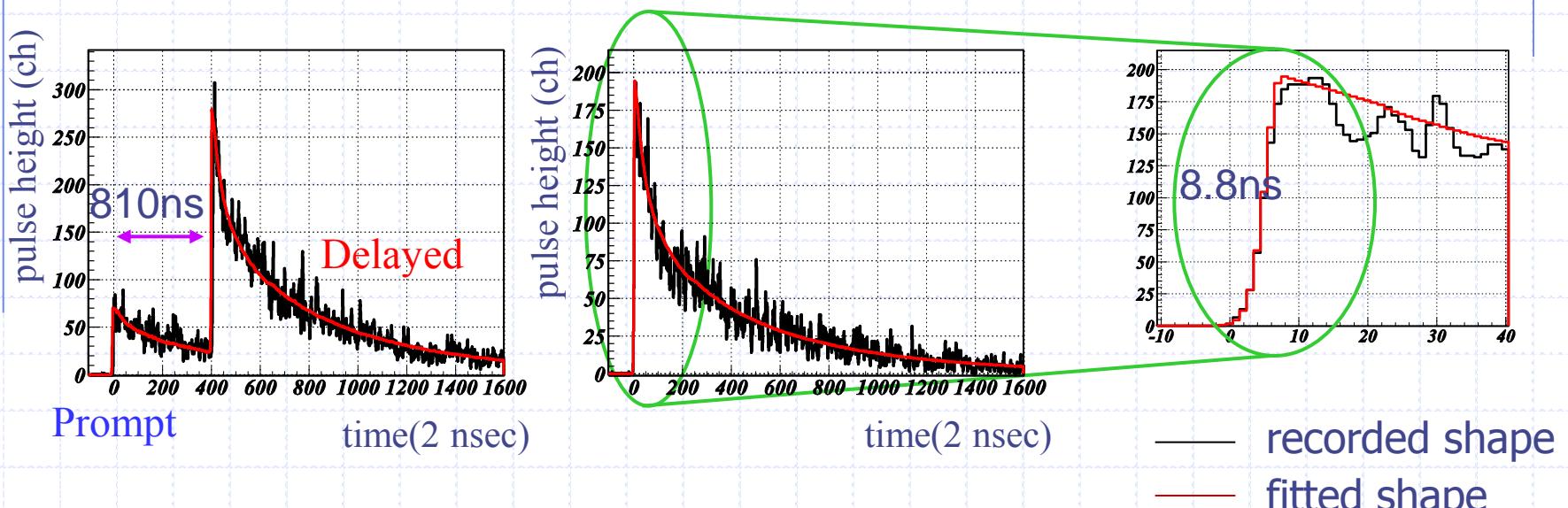
$^{212}\text{Bi}$  (Th)



$$\begin{aligned} E_{\max} &= 5.8 \text{ MeV (U)} \\ &5.3 \text{ MeV (Th)} \end{aligned}$$

# Rejection of Double Pulse(DP)

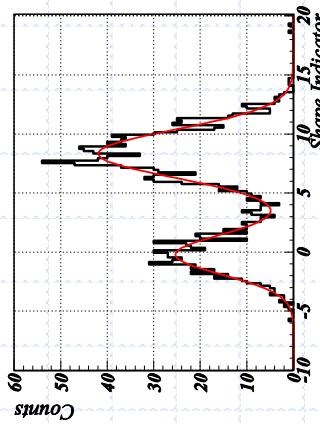
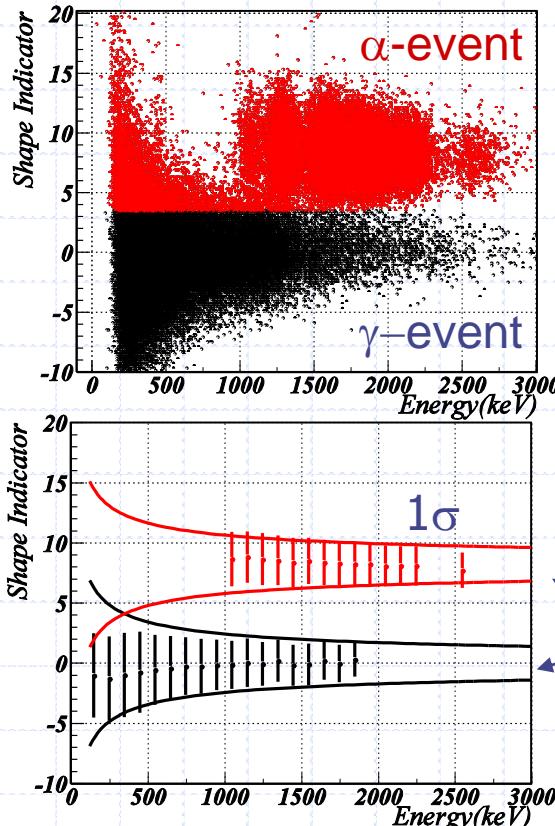
## Typical Pulse Shapes



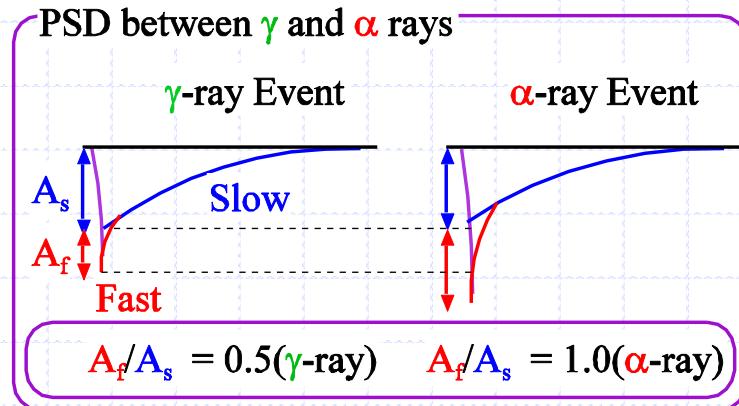
# Pulse Shape Discrimination

## ◆ Pulse Shape discrimination

- Shape Indicator (PRC **67**(2003) 014310)

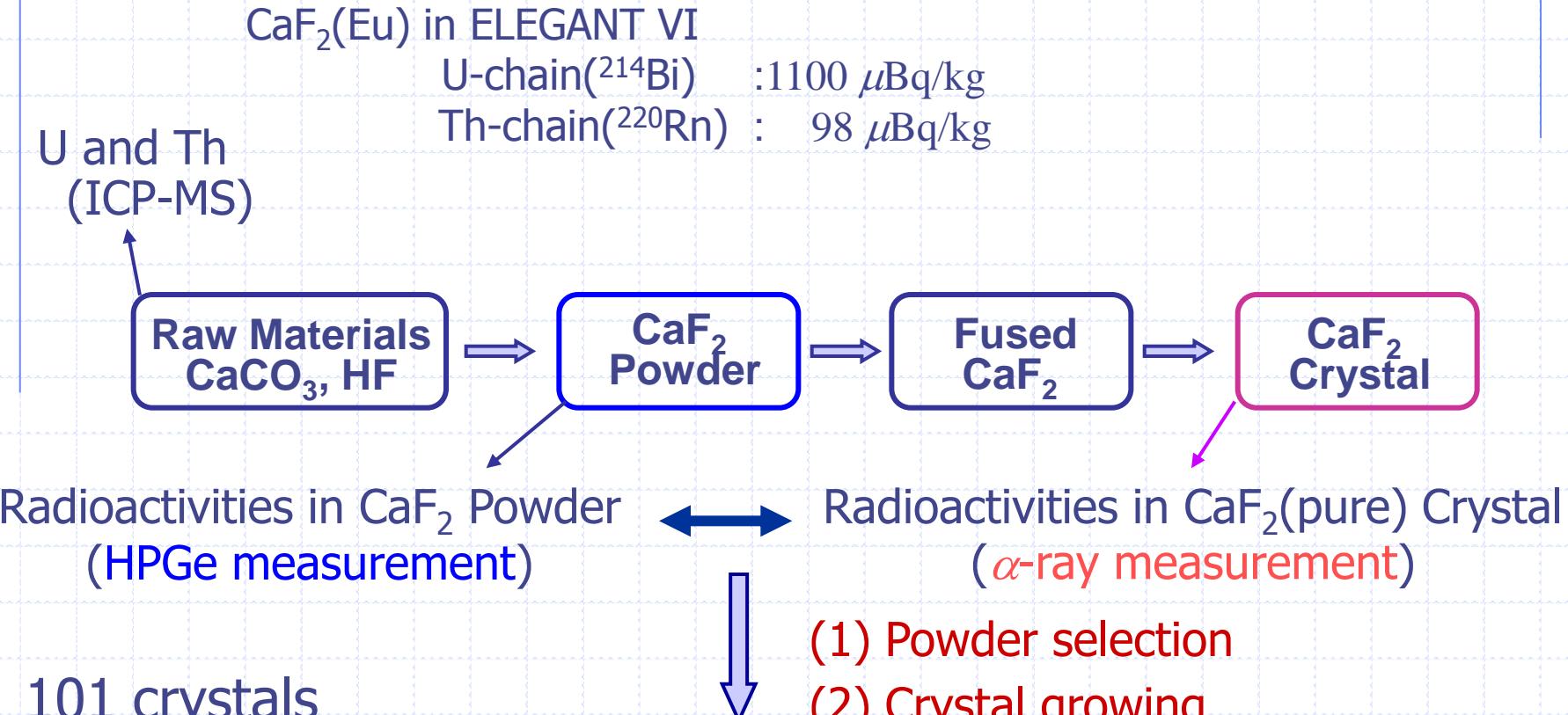


Difference in decay shape between  $\alpha$  and  $\gamma$  rays



mean value:  
no energy dependence (>1 MeV)

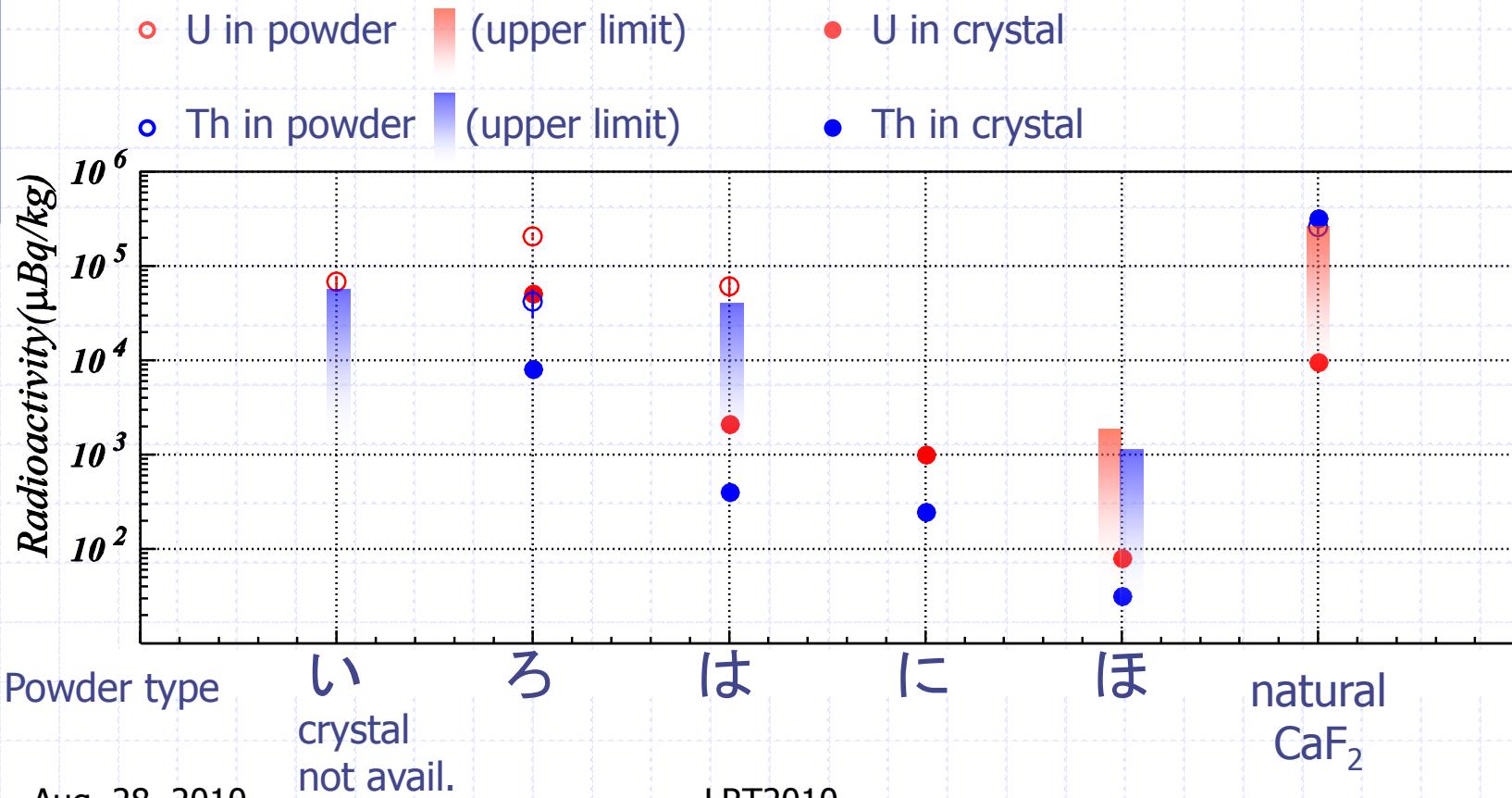
# Development of High Purity $\text{CaF}_2$ Crystals



# (1) Powder selection

◆ Measurements before (powder) and after crystallization

Oken co., Ltd.

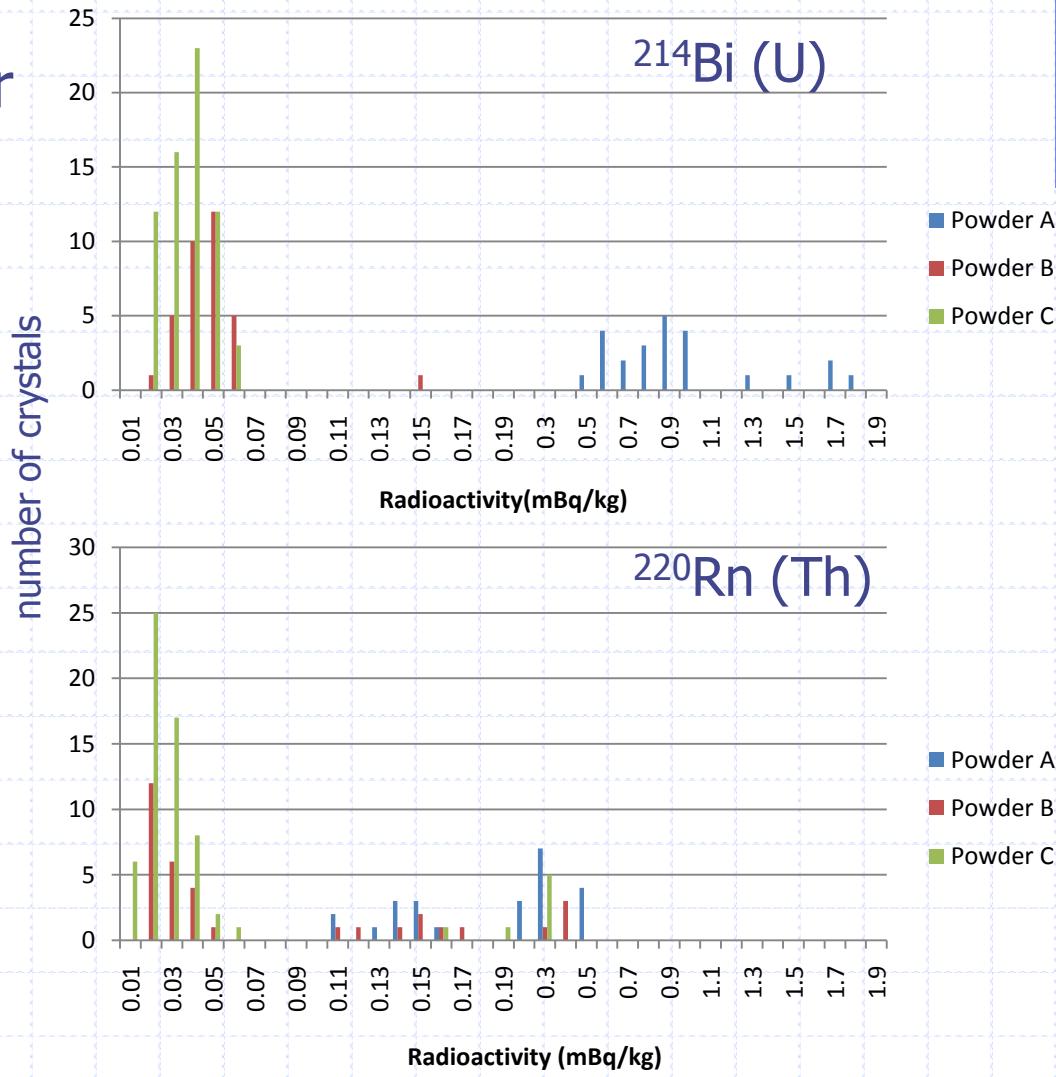




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# Powder type

- 3 types of  $\text{CaF}_2$  powder
  - Powder A  
24 crystals
  - Powder B  
34 crystals
  - Powder C  
66 crystals

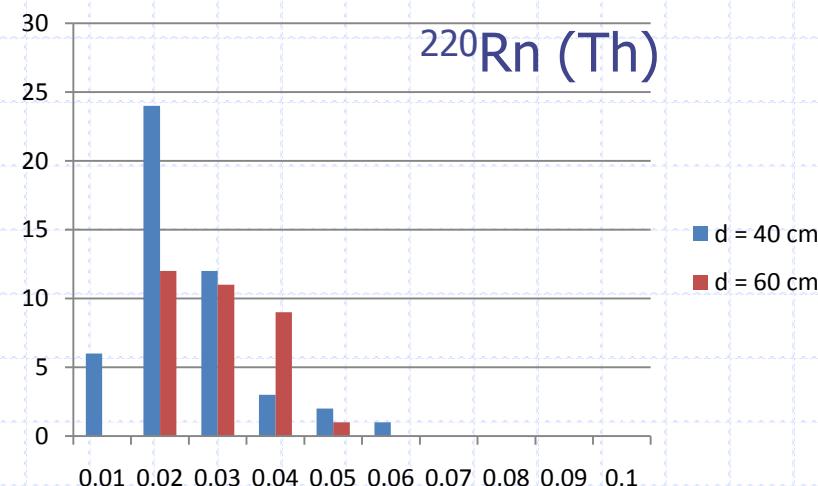
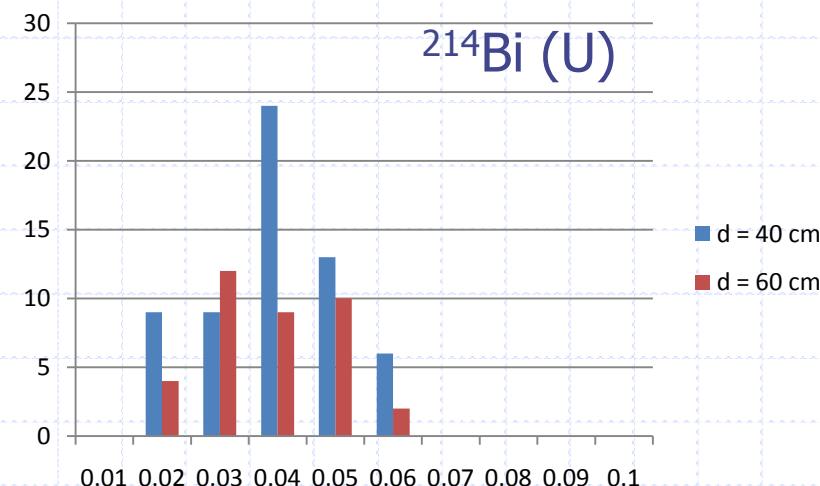




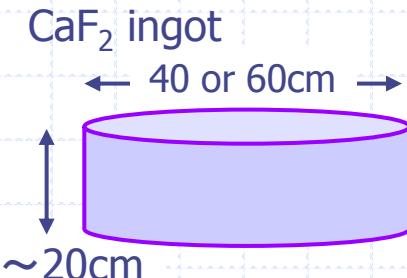
Candles

## (2) Crystal growing

- ◆ Two types of melting pod
  - $d = 40$  or  $60$  cm



→ There is no big difference



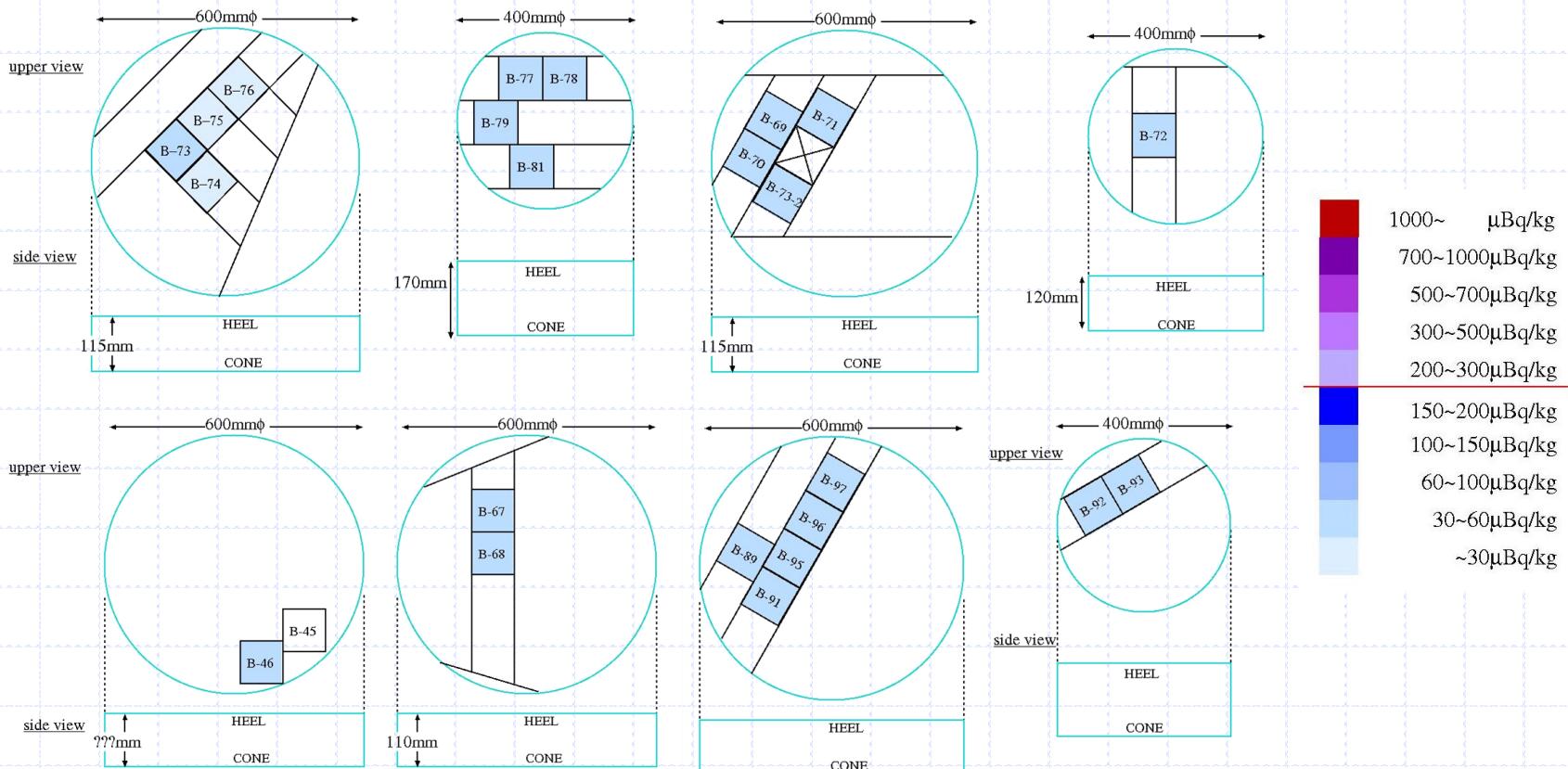


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# Position dependence

◆  $^{214}\text{Bi}$ (U-chain)

→ no position dependence



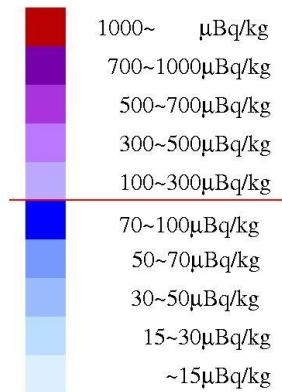
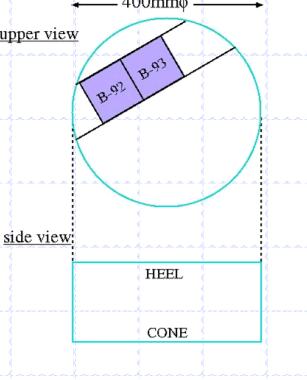
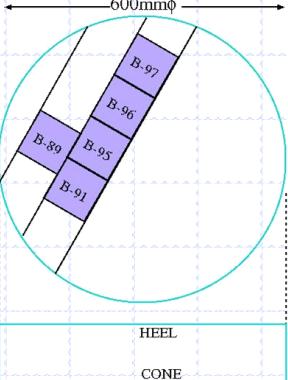
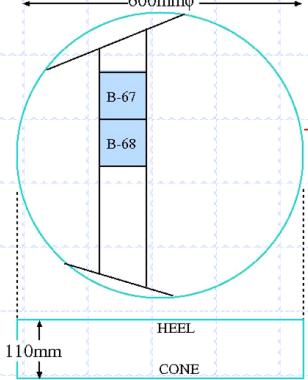
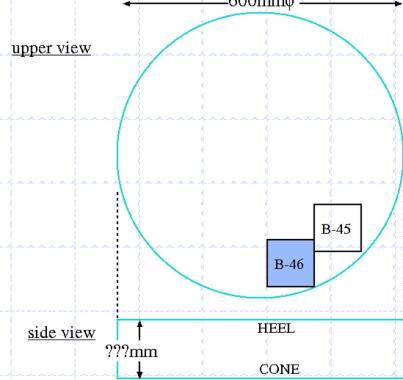
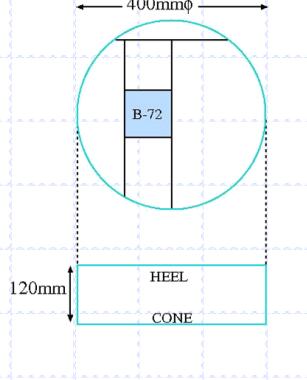
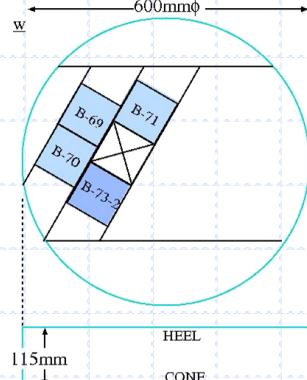
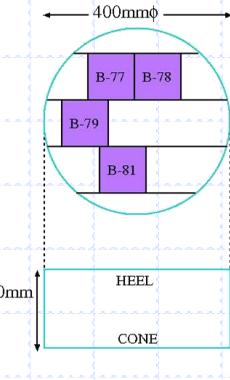
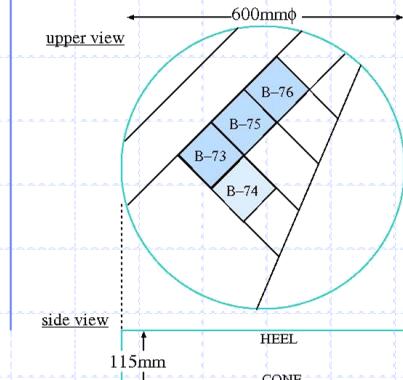


Candles

# Position dependence

◆  $^{220}\text{Rn}(\text{Th-chain})$

no position dependence

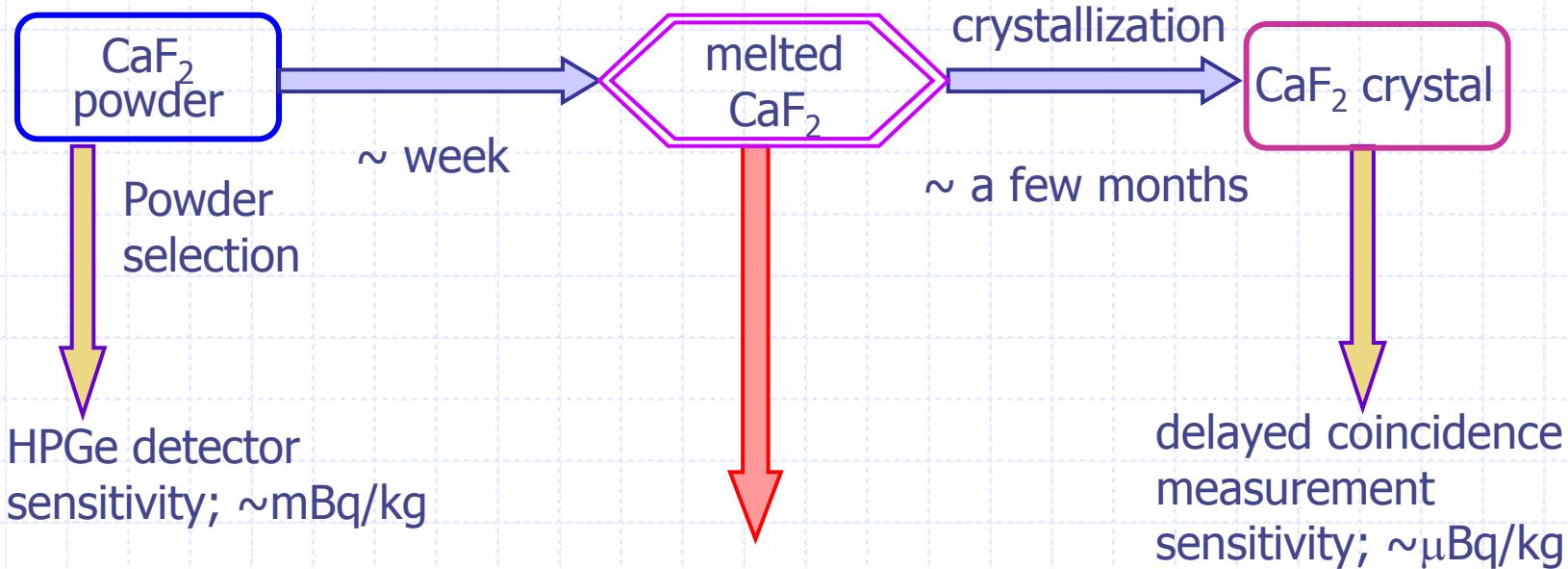




Candles

# R&D (1)

- Purity measurement using melted  $\text{CaF}_2$



- ◆ Can we measure the radioactivity of melted  $\text{CaF}_2$ ?

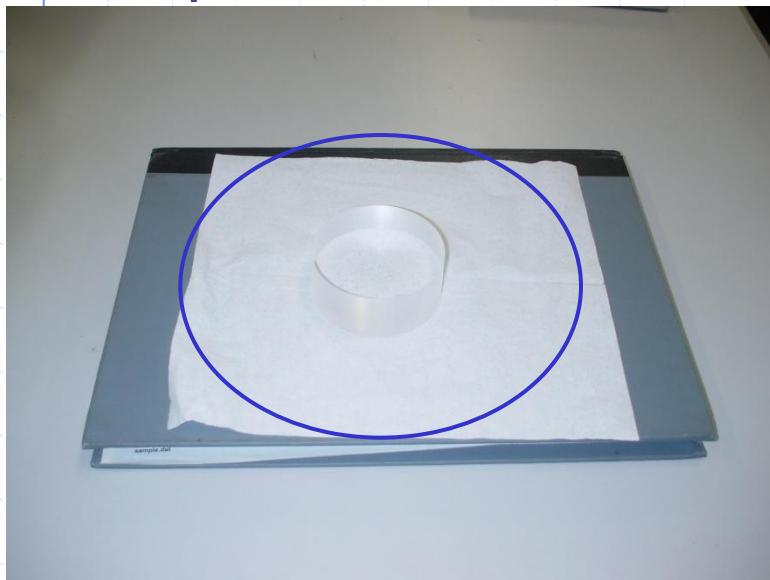
- If scintillated, we can use delayed coincidence technique.
  - high sensitivity in short time



Candles

# Melted CaF<sub>2</sub> samples

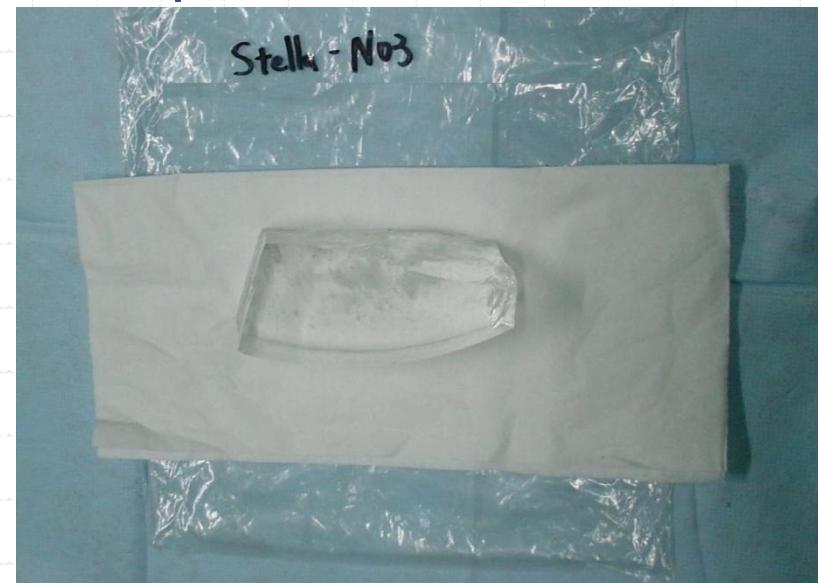
Sample #1



367.5g,  $\phi 7 \times t3$  cm<sup>3</sup>

from pure CaF<sub>2</sub> powder

Sample #2



341.7g

High radioimpurity  
CaF<sub>2</sub> powder

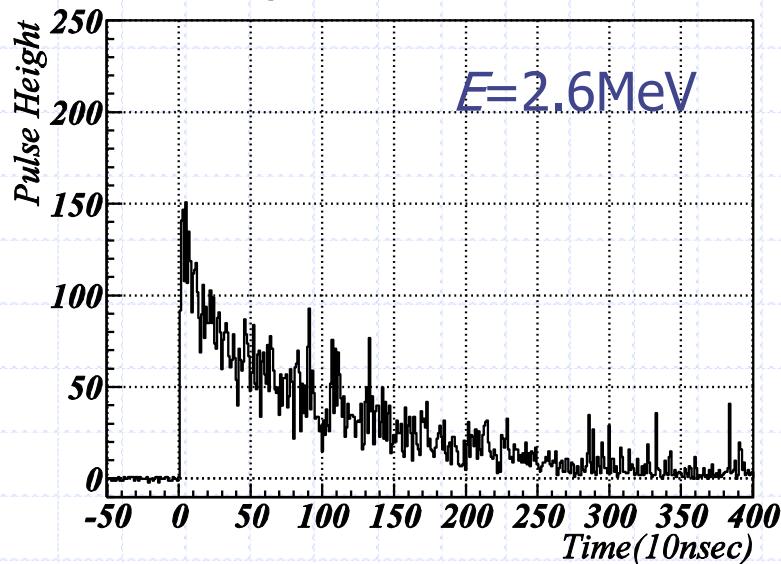


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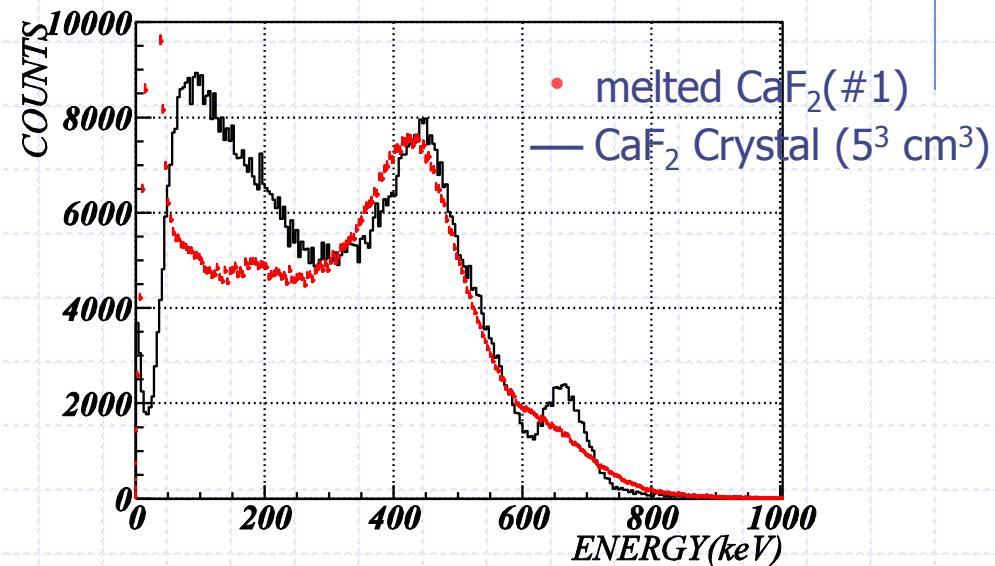
Stella Chemifa Corp.

# Scintillation measurement

Pulse shape



Energy spectra ( $^{137}\text{Cs}$ )



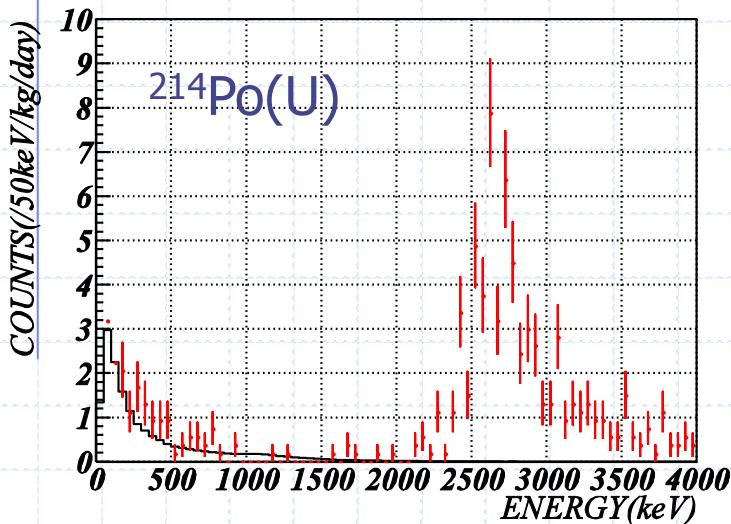
- Weak scintillation light observed
- poorer energy resolution, but enough for radioactivity measurement

# Radioactivity measurement



Stella Chemifa Corp.

## Sample #1

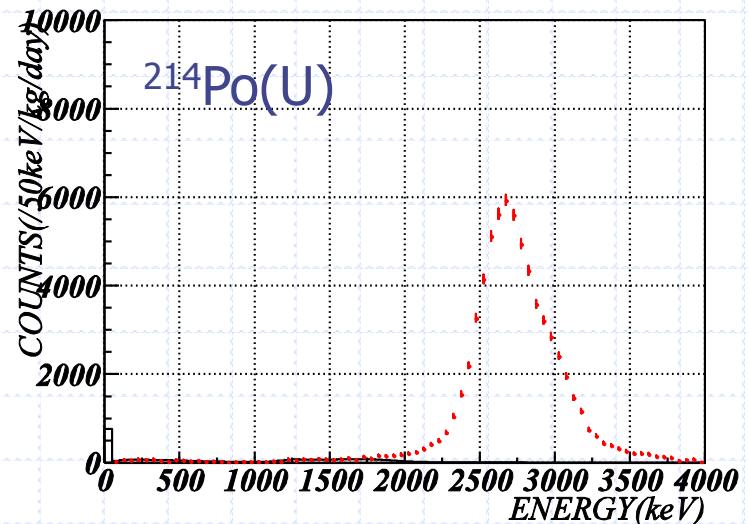


$^{214}\text{Po}(\text{U})$  :  $0.87 \pm 0.05\text{mBq/kg}$

$^{216}\text{Po}(\text{Th})$  :  $0.083 \pm 0.020\text{mBq/kg}$

meas. time: 14.5 days

## Sample #2



$^{214}\text{Po}$  :  $927 \pm 6\text{mBq/kg}$

$^{216}\text{Po}$  :  $< 9.1\text{mBq/kg}$

1.0 day

# comparison with ICP-MS

## ◆ Delayed coinc.

### Sample #2

$^{214}\text{Po}(\text{U})$ :  $75.0 \pm 0.5$  ppb  
 $^{212}\text{Po}(\text{Th})$ : <2.2 ppb



### ICP-MS

U: 80 ppb  
 Th: <20 ppb

### Sample #3

$^{214}\text{Po}(\text{U})$ :  $100.1 \pm 0.7$  ppb  
 $^{212}\text{Po}(\text{Th})$ : <3.3 ppb

U: 100 ppb  
 Th: <20 ppb

### sensitivity

$^{214}\text{Po}(\text{U})$ : ~0.001 ppb  
 $^{212}\text{Po}(\text{Th})$ : ~0.002 ppb

### sensitivity

U	0.2 ppb
Th	0.3 ppb

# R&D (2)

## ◆ Rinse the powder with HNO<sub>3</sub> (3 wt%)

Oken co., Ltd.

- same powder (type, lot)
- contamination measurement was done after crystallization

without rinsing

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



with rinsing

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

# Summary

- ◆ CANDLES project
  - Study of  $^{48}\text{Ca}$  double beta decay
- ◆  $\text{CaF}_2$ (pure) scintillation crystal
  - Material ( $\text{CaF}_2$  powder) selection is quite important
  - No significant difference in radioactivity measurement with two types of ingot size
  - No position dependence in radioactivity measurement for each ingot
  - Melted  $\text{CaF}_2$  can be used to measure the radioactivity in the  $\text{CaF}_2$  powder
- ◆ CANDLES III(U.G.) @Kamioka
  - Under construction
  - Expected BG: 0.18 ev/year

# CANDLES Collaboration

- ◆ Osaka U. (大阪大学)  
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