Purification of CaF₂ crystal for double beta decay experiment



Ken–Ichi Fushimi For CANDLES collaboration

CANDLES project outline

- Aim : search for neutrino-less double beta decay.
- Target nucleus : ⁴⁸Ca (350 g total)
 - Q-value = 4.27 MeV, the highest value.
 - Almost background free.
- Present sensitivity
 - 5.6×10²² yr (90% C.L.)
 - 130.4 days
- Serious background
 - Radioactive impurity in CaF₂.
 - Beta and gamma rays from $^{\overline{2}08}$ TI.
 - External gammas by (n,γ) .

S.Ajimura et al. Phys. Rev. D <u>103</u>, 092008 (2021)





Main background origin

- ²⁰⁸TI in CaF₂ crystal
 Q_β = 5.001 MeV
 β+γ in the crystal
 Serious BG
- Present CaF₂ crystal contains 2~60 μBq/kg of Th-series.



Purification of CaF₂ crystal

- Difficulties: CaF₂ is not water soluble.
- We cannot apply the method for NaI.
 - NaI(TI) was successfully purified by PICOLON group.
 - K.Fushimi et al. PTEP 2021, 043F01 DOI: 10.1093/ptep/ptab020
- Purification of surrounding material
 - Crucible selection
 - Crystallization procedure
 - Position dependence of RI concentration

Crucible selection

- Normal graphite
- High purity graphite without polish \leftarrow The best value.
- Polished high purity graphite
- Melting and coagulation were done in IMR Tohoku Univ.









Position dependence in CaF₂

- Segregation in long crystal
- The effective purification method.
- Long crucible was used.







RI measurement

- Th-series contamination was measured by delayed coincidence.
- Measurement : Kamioka underground lab-A.





Shield: Cu 5 cm + Pb I 0 cm

Crystal is attached on a PMT.

Analysis: ²¹⁴Bi-²¹⁴Po (U-ser.) & ²²⁰Rn-²¹⁶Po (Th-ser.)



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Results

Sample	U-ser.(²¹⁴ Po) mBq/kg	Th-ser.(²¹⁶ Po) mBq/kg
High purity polished crucible	2.45 ± 0.25	2.20 ± 0.28
High purity unpolished crucible	3.37±0.18	2.53±0.18
High purity unpolished crucible: upper	2.14±0.19	3.33±0.18
High purity unpolished crucible: lower	0.88±0.09	1.38±0.11

Significant reduction in the lower part of the crystal. Purification by zone refining worked well !

Discussion and prospect

- No significant difference of crucible.
 - We chose to use high-purity crucible by conservative decision.
- Zone refining worked.
 - The concentration reduced by 1/2.
 - To be confirmed for reproducibility.
- ICP-MS measurement for future development
 - Quick and precise measurement of raw materials.
 - Measurement of Ra, Th, and U in $CaCl_2$ and other materials.
- CaCl₂ is water soluble → Same purification method for NaI is applicable.