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Ultra-purification and mass-production of NaI powder for COSINE-200

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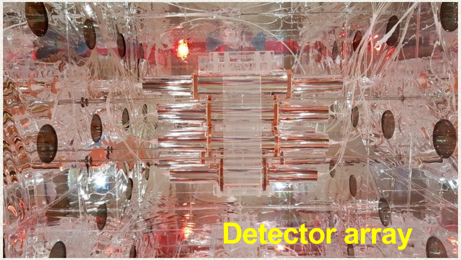
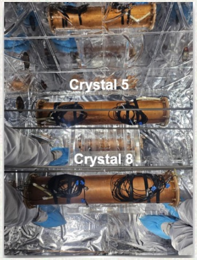
COSINE experiment overview

The COSINE experiment is searching for dark matter using ultra-low background NaI(Tl) scintillating crystals to verify the DAMA/LIBRA’s claim via the same target material.

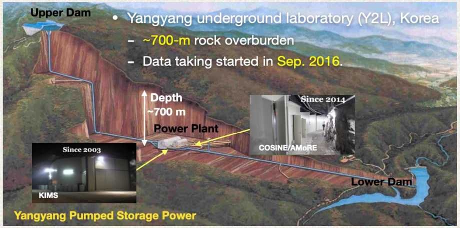


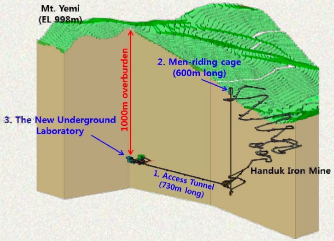


COSINE-100
2016 – 2023

COSINE-100Upgrade

COSINE-200





Yemilab
EST. 2022
Less Muon
Better Flavor

Towards COSINE-200

01. <u>NaI powder purification</u> – In-house technology required

- The NaI powder for COSINE-100 crystal was selected by the producer, Alpha-Spectra Inc. company.
- The crystal was found to have three times higher intrinsic contamination than the DAMA/LIBRA crystal.
- The main background sources are internal ^{210}Pb, ^{40}K, and cosmogenic ^{3}H.

Eur. Phys. J. C (2021) 81:837

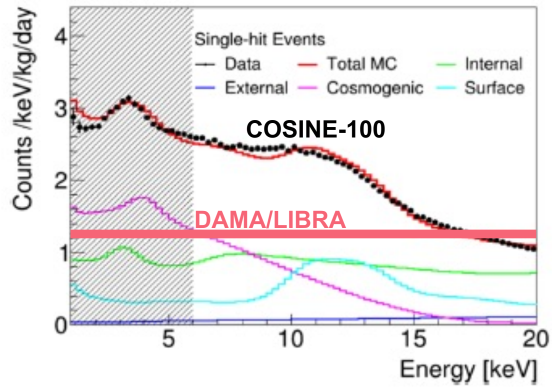


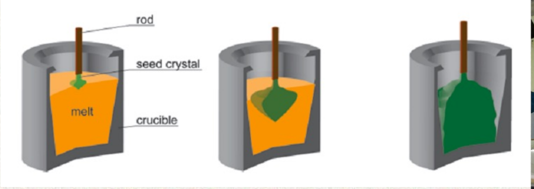
Fig. 9 The low-energy spectra of single-hit events averaged for the five crystals. The measured energy spectrum after efficiency corrections [23] is compared with the total of the simulations. The range of 1–6 keV in the MC spectrum is extrapolated from the modeling

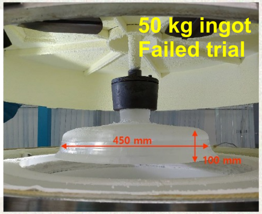
Towards COSINE-200

02. Crystal growing – In-house technology required

- 106 kg COSINE-100 crystal was produced by Alpha-Spectra Inc. company.
- A small crystal grower was used for proof of principle.
- The growing technology using full-size crystal grower is under development

Kyropolous growers with quartz crucible



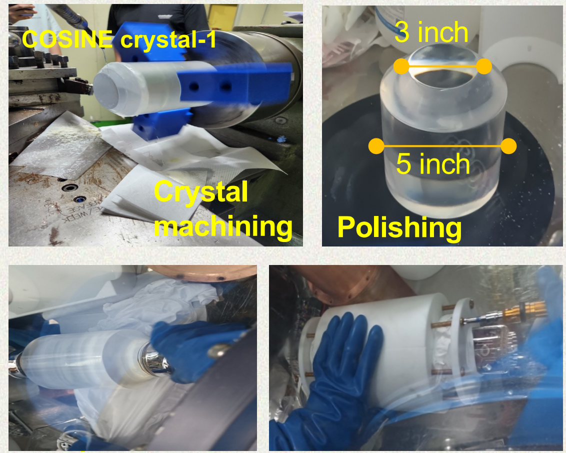


Towards COSINE-200

NIMA 981 (2020) 164556

03. Crystal machining and polishing – In-house technology required

- 106 kg COSINE-100 crystal was produced and encapsulated by Alpha-Spectra Inc. company.
- 40% increased light yield was achieved with the COSINE-100U setup



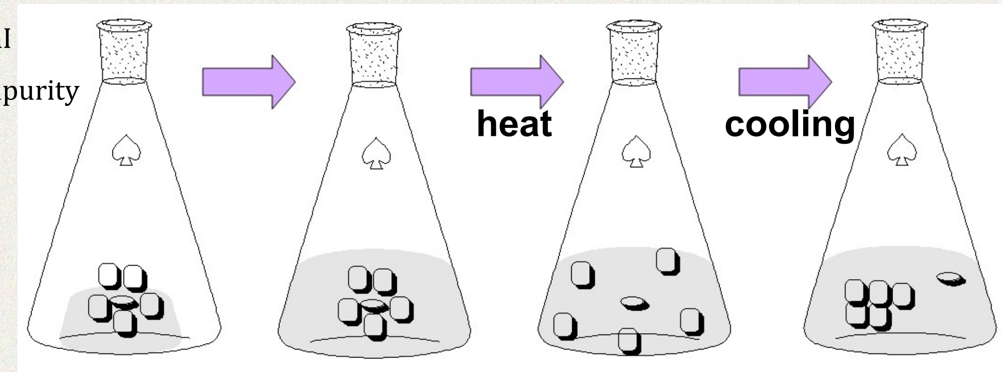
04. Detector assembly

Astropart. Phys. 141, 102709 (2022)

Purification via fractional recrystallization

 NaI

 Impurity



1. Selection of an appropriate solvent.

2. Add just enough solvent to dissolve solid.

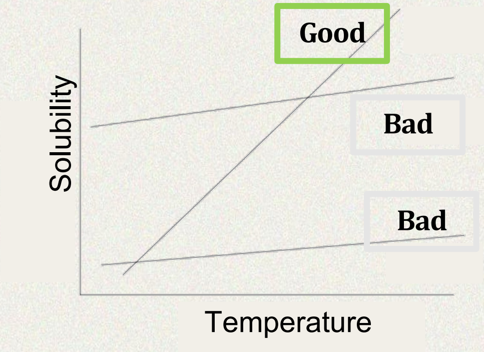
3. Evaporate some solvent to make an oversaturated solution.

4. Cool down slowly to get the pure crystals leaving impurities in solution

 NaI

 210 Pb | 40 K | 226 Ra | 232 Th | 238 U

Purification via recrystallization from aqueous solutions



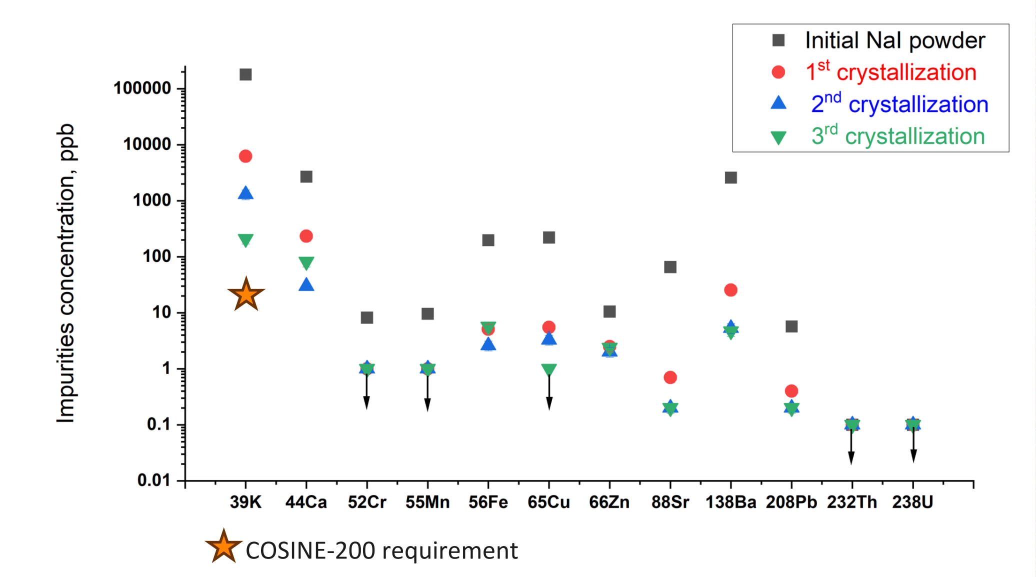
NaI solubility
(g/100ml of solvent)
water: 159.7 (0°C)
water: 179.3 (20°C)
water: 205 (40°C)
water: 257 (60°C)
water: 296 (80°C)

NaI

^{210}Pb ^{40}K ^{226}Ra ^{232}Th ^{238}U

	^{nat}K	^{232}Th	^{238}U
DAMA/LIBRA powder [NIMA 592 (2008) 297-315]	<100 ppb	20 ppt	20 ppt
COSINE-200 requirements	<20 ppb	<10 ppt	<10 ppt

Lab scale _ Technical grade powder



Decontamination factor

J. Rad. Nucl. Chem. 317, 1329 (2018)

Material	^{39}K	^{44}Ca	^{52}Cr	^{55}Mn	^{56}Fe	^{65}Cu	^{66}Zn	^{88}Sr	^{138}Ba	^{208}Pb
1st crystallization cycle	28.7	11.5	> 8.2	> 9.6	38.7	40.3	4.0	93.9	101.6	14.3
2nd crystallization cycle	137.9	> 90	> 8.2	> 9.6	76.0	67.2	5.3	328.5	489.0	28.5
3rd crystallization cycle	857.1	33.0	> 8.2	> 9.6	34.6	> 221.6	4.4	328.5	551.4	28.5

Lab scale _ Astro & Crystal grades

Material	Astro grade, 99.999 + %		Crystal grade, 99.99(5) %	
	Initial	Purified	Initial	Purified
Unit	ppb			
^{39}K	4.5	< 1	45.1	6.0
^{44}Ca	16.0	< 20	94.6	30.4
^{52}Cr	19	< 1	23.7	< 1
^{55}Mn	1.7	< 1	< 1	< 1
^{56}Fe	110.1	< 3	34.6	3.9
^{65}Cu	1.7	< 1	11.5	< 1
^{66}Zn	3.8	< 3	9.1	< 3
^{88}Sr	0.3	< 0.3	0.9	< 0.3
^{138}Ba	0.6	< 0.3	7.1	0.6
^{208}Pb	0.9	0.4	3.3	0.8
^{232}Th	< 0.1	< 0.1	< 0.1	< 0.1
^{238}U	< 0.1	< 0.1	< 0.1	< 0.1

J. Rad. Nucl. Chem. 317, 1329 (2018)

- Astro-grade powder satisfies the COSINE-200 requirements on purity but goes out of budget.
- Crystal-grade powder requires just one cycle of recrystallization to reach the purity of Astro-grade powder.
- <20 ppb of K in purified powder is achievable.
- Pb reduction is still noticeable at ppb and ppt level.

Crystal grade + single crystallization = Astro grade

Mass production facility

- In-house designed and Commissioned in 2019 at CUP

Mass production facility

fphy.(2023) 1142849

Feed tank

Mixing tank

Filter unit

Receiver tank 1, 2

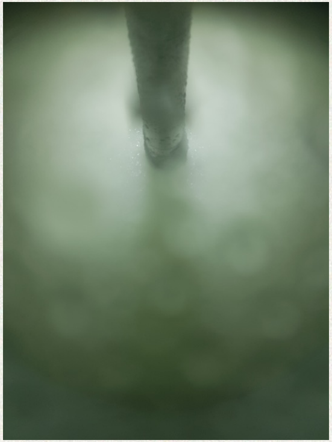
Conical dryer

Controller

Mass production facility

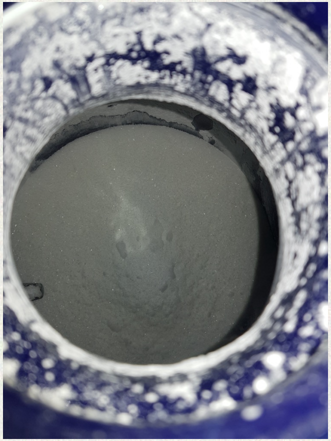
fphy.(2023) 1142849

Preparing of oversaturated solution at 100 ˚C



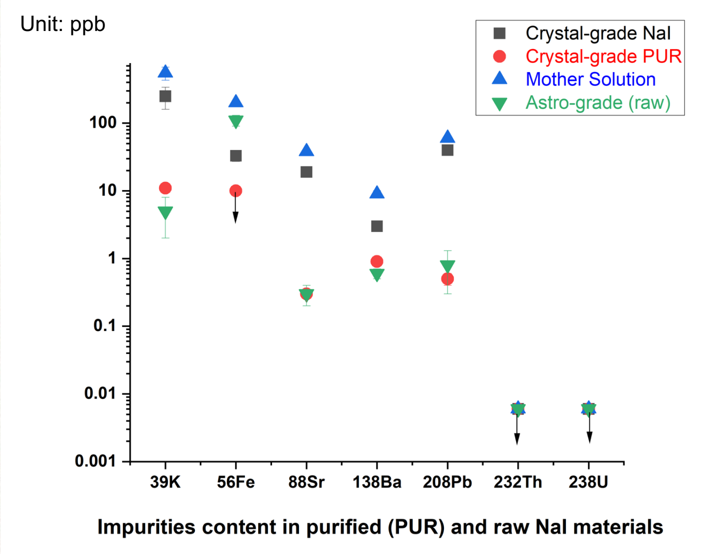
Recrystallized crystals and mother solution

Filtering out and washing the crystals



Dry the crystals in the conical dryer

Purification of Crystal-grade NaI (CG-NaI)

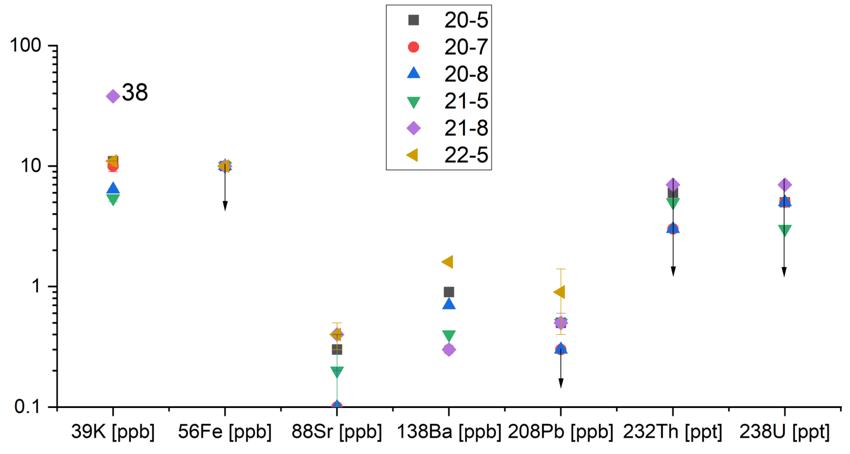


Proof of principle confirmation in mass-scale production:

- The purity of Astro-grade (AG-NaI) powder was achieved with single re-crystallization of the Crystal-grade (CG-NaI) NaI.
- The Mother Solution (MS) remaining after CG-NaI recrystallization is similar to the purity of the raw CG-NaI. The MS could be recycled.
- Th and U in all powders were below 6 ppt.

HPGe meas. Crystal-grade PUR, mBq/kg			
$^{226}Ra(^{238}U)$	^{40}K	^{228}Ac	^{228}Th
< 0.56	< 4.04	< 0.96	< 0.85

Purification of Crystal-grade NaI (CG-NaI)

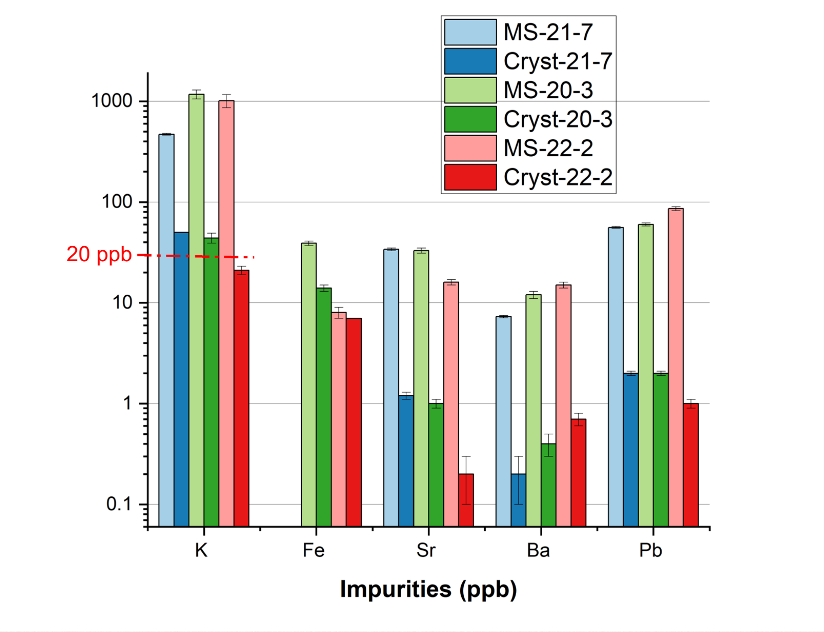


Impurities content in different NaI batches produced at CUP

We keep the balance between the crystallization rate (<50%) and purity (<20 ppb K)

	K [ppb]	Fe [ppb]	Sr [ppb]	Ba [ppb]	Pb [ppb]	Th [ppt]	U [ppt]
Overall range of impurities within 35-44% crystallization rate	5.1 / 12	<7 / <10	0.1 / 0.5	0.2 / 1.8	<0.3 / 1.4	<3 / <10	<3 / <10

Mother Solution (MS) recycling



Reduction of impurities in MS. Different experimental runs.

35% Crystallization rate

To reach purity level of ^{nat}K < 20 ppb

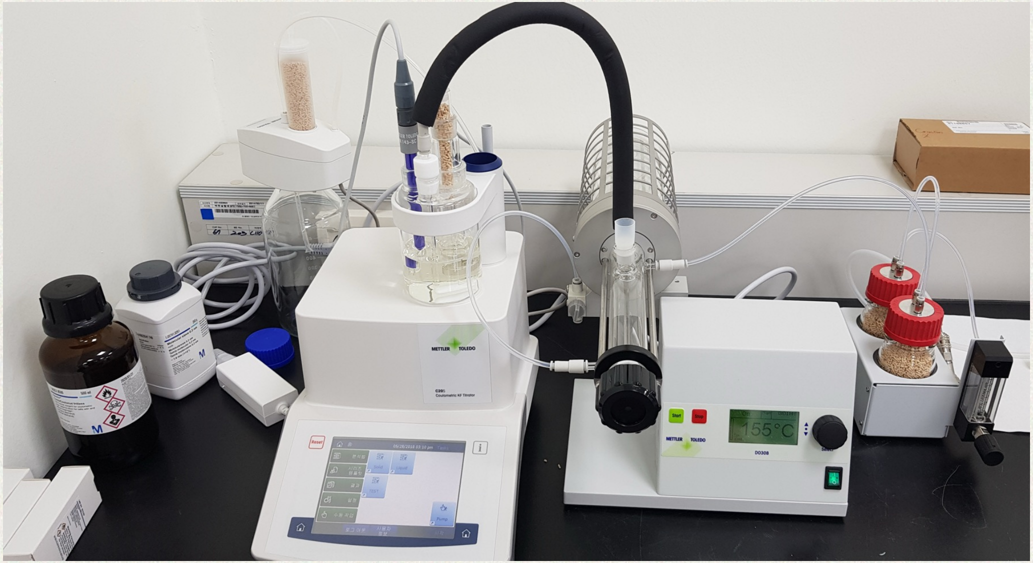
Maximum 1 ppm K in MS is allowed

To reach the purity in one crystallization cycle

^{232}Th and ^{238}U

Th and U levels in MS and produced crystals are < 7 ppt

Water content measurement



- Karl-Fisher Titration, ppm to % level of moisture in the powder
- The required level for successive crystal growing is < 0.1%
- Excessive moisture in the powder may cause damage to the quartz crucible and adversely affect the crystal growth

Crystals grown at CUP

	Powder used	^{nat}K [ppb]	^{210}Pb [μBq/kg]	^{232}Th (^{216}Po) [μBq/kg]	^{238}U average [μBq/kg]	Comment
NaI-025	AG-NaI	684 ± 100	3.8 ± 0.3	< 6	26 ± 7	Refractories were replaced
NaI-034	AG-NaI	< 62	0.05 ± 0.09	35 ± 5	51 ± 7	
NaI-035	AG-NaI	< 42	0.01 ± 0.02	7 ± 2	11 ± 4	Best purity was observed
NaI-036	AG-NaI	< 53	0.42 ± 0.27	< 20	451 ± 48	Contamination in the growing process
NaI-037	PUR-NaI	8.3 ± 4.6	0.38 ± 0.10	< 3.3	< 25	

Eur. Phys. J. C (2020) 80:814

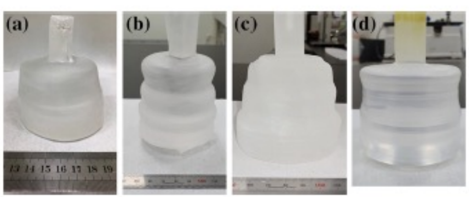
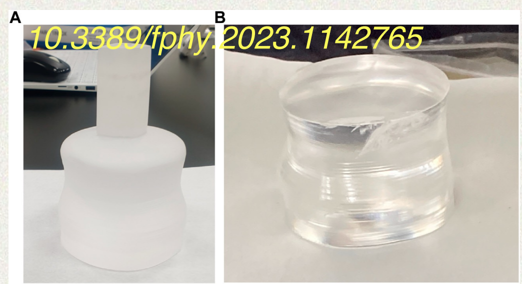
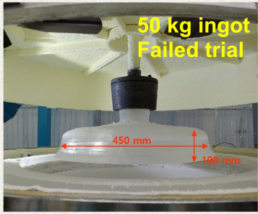


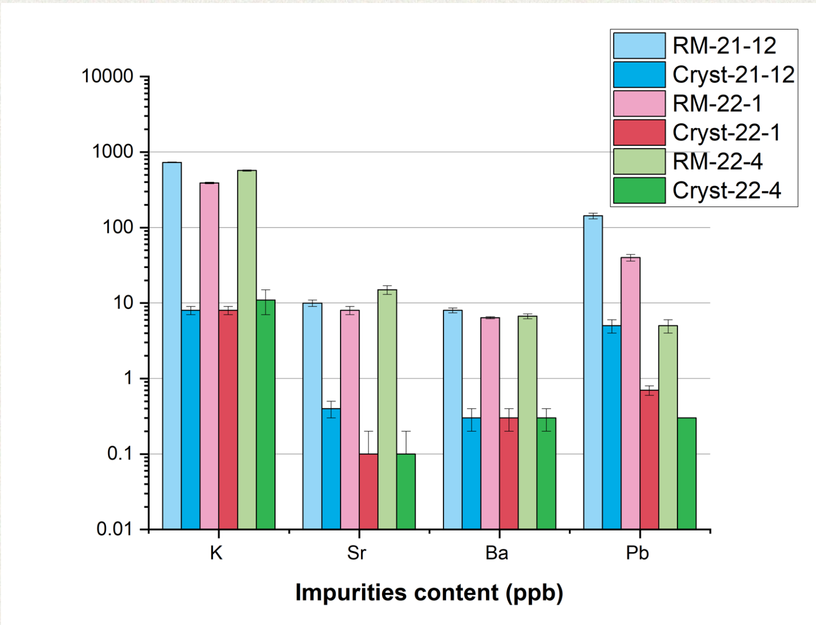
Fig. 3 Four NaI(Tl) crystal ingots produced by the small grower : **a** NaI-025, **b** NaI-034, **c** NaI-035, and **d** NaI-036



NaI-037 crystal. (A) Crystal ingot and (B) polished crystal



Residual Melt (RM) recycling



Reduction of impurities in RM. Different experimental runs.

CG-NaI powder

To establish the large size crystal growing technology

50% crystallization rate

To reach the purity I one crystallization cycle

^{232}Th and ^{238}U

Th and U levels in MS and produced crystals are < 7 ppt

Reported purity for recently grown NaI(Tl) crystals

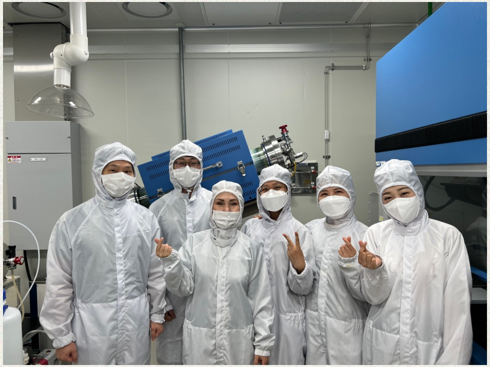
Unit: μBq/kg

	COSINE #NaI-035 [1]	COSINE #NaI-037 [2]	COSINE-100 #6 [1]	DAMA [3]	SABRE #33 [4]	PICOLON #85 [5]	ANAIS-112 [6]
Ingot mass, kg	0.6	0.7	12.5	9.7	3.4	1.3	112.5
NaI powder / growing facility	AG-NaI / CUP	CG-PUR / CUP	Alpha Spectra comp.	Saint Gobain comp.	AG-NaI / RMD comp	In-house tech.	Alpha Spectra comp
^{40}K	< 1300	260 ± 140	520 ± 80	<620	150 ± 20	<600	700 – 1330
^{210}Pb	10 ± 20	380 ± 100	1870 ± 90	10 – 30	461 ± 5	<5.7	700 – 3150
^{232}Th	7 ± 2	< 3	2.5 ± 0.8	2 - 31	1.6 ± 0.3	0.3 ± 0.5	0.4 – 4.0
^{238}U	11 ± 4	< 24.4	< 0.25	8.7 - 124	6.0 ± 0.6	1.0 ± 0.4	3 – 10

[1] Eur.Phys.J.C. (2020) 80:814; [2] 10.3389/fphy.2023.1142765; [3] NIMA 592 (2008) 297-315; [4] Eur. Phys. J. C. (2022) 82:12; [5] Present status of PICOLON project. ~Purity of NaI(Tl) and background ~. Kenta Kotera. Tokushima University. @ DMNet. 2024; [6] Eur. Phys. J. C 79, 412 (2019).

Summary

- Methods of NaI purification and recycling were developed and performed at CUP.
- Mass-scale purification facility was established and commissioned at CUP. The maximum production capacity is 70 kg of powder in two weeks.
- The purification facility supports the crystal growing trials via developing and performing the successive recycling technology for the residual melt.
- 400 kg of pure NaI powder had been produced at CUP.
- Further development of the purification of TlI could be introduced if needed.



Thank you for your attention!