CENTER FOR UNDERGROUND PHYSICS

Institute for Basic Science 기초과학연구원 ibs

Material PURification and LRT at CUP

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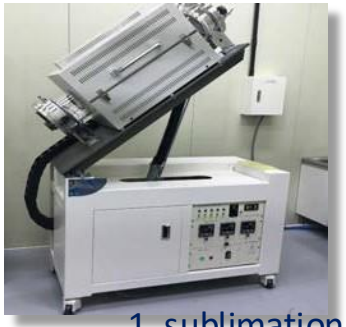
$^{100}MoO_3$ for AMoRE-II

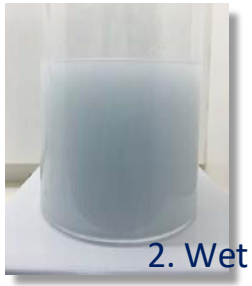
01

$^{100}MoO_3$ raw material selection and purification

1. In-house designed sublimation apparatus: JINST 14(11):T11002-T11002
2. Wet chemistry: co-precipitation and membrane filtration. In-house purified co-precipitation agent.
3. Purification capacity at CUP: 5 kg/month
4. Recovery efficiency for the process 99%: Frontiers in Physics 11
5. Irrecoverable losses: 1%
6. Purity of the produced powder is controlled with ICP-MS and HPGe array at CUP.



1. sublimation under low vacuum



2. Wet chemistry

4. final $^{100}MoO_3$ powder

3. Synthesis of ammonium polymolybdate powder and its annealing

$^{100}MoO_3$ for AMoRE-II

02

ICP-MS assay of $^{100}MoO_3$ raw and purified powders at CUP

Element	Al, [ppb]	K, [ppb]	W, [ppb]	Sr, [ppb]	Ba, [ppb]	Pb, [ppb]	Th, [ppt]	U, [ppt]
$^{100}MoO_3$ raw powder (range for all received lots)	500 - 2100	300 - 1600	10 - 1350	2 - 80	8 - 20	3 - 9	30 - 150	40 - 280
CUP PUR $^{100}MoO_3$ powders (range for all produced powders)	<100 - 700	<500 - 1100	<50 - 700	<0.2	<4	<0.5	<7	<7
DF	1 – 20	1—3		10 – 400	2 – 5	6 – 18	3 – 20	5 – 40

*DF stands for Decontamination Factor

$^{100}MoO_3$ for AMoRE-II

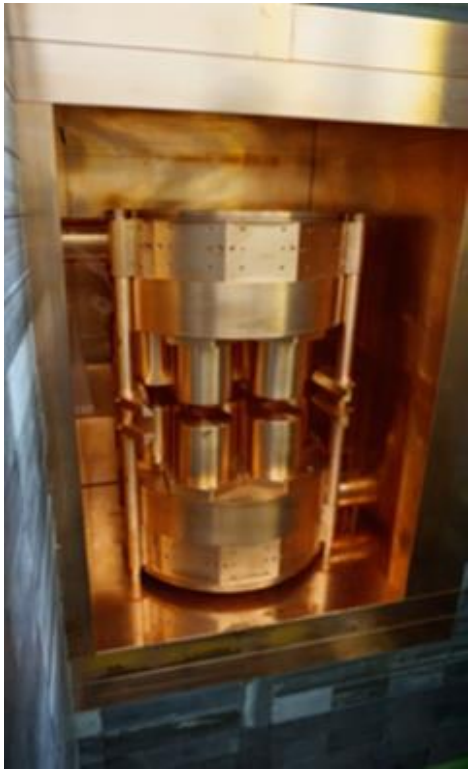
03

HPGe array measurement for 13 kg $100MoO_3$, raw and purified

HPGe array measurement of 13 kg of $^{100}MoO_3$ purified at CUP

μBq/kg	^{228}Ac	^{228}Th	^{226}Ra	^{40}K
$^{100}MoO_3$ raw powder (range for all received lots)	260 ± 50	210 ± 50	260 ± 50	8500 ± 1400
CUP PUR $^{100}MoO_3$ powders (range for all produced powders)	<27	<16	110 ± 30	1700 ± 340
DF	9	13	2	5

*DF stands for Decontamination Factor



The array of 14 HPGe detectors with inner copper shielding of ARRAY detector

Li_2CO_3 selection for AMoRE-II

01

ICP-MS assay of various commercial Li_2CO_3 powders at CUP

	^{40}K	^{208}Tl	^{226}Ra (^{214}Bi)	^{228}Ac
Alfa Aesar, Puratronic®, 99.998%				
2016	9 ± 3.4	0.41 ± 0.22	0.95 ± 0.22	1.4 ± 0.64
2021 1300 USD/kg	26.5 ± 7.1	<3.8	28 ± 2	<5.6
2017 [26]	≤42	n/d	705 ± 30	12 ± 4
NRMP powders				
TU 6-09-3728-83	<11.5	<1.8	<1.8	<1.7
Pharma grade	<66.4	9.1 ± 2.6	2,730 ± 137	108.2 ± 9.3
Tech. Grade LiOH	<5.1	<3.8	730 ± 20	28 ± 4
Li_2CO_3 from Chinese market				
A, 5N	<16.6	<1.3	57.4 ± 3.2	6.3 ± 1.3
B, 5N	<10.9	11.4 ± 1.4	295 ± 15	12 ± 2
C, 5N	<86.7	185 ± 14	10,850 ± 540	435 ± 35

Values and upper limits are given at 95% confidence interval. The "n/d" stands for no data.

Unavailable

Yellowish crystals

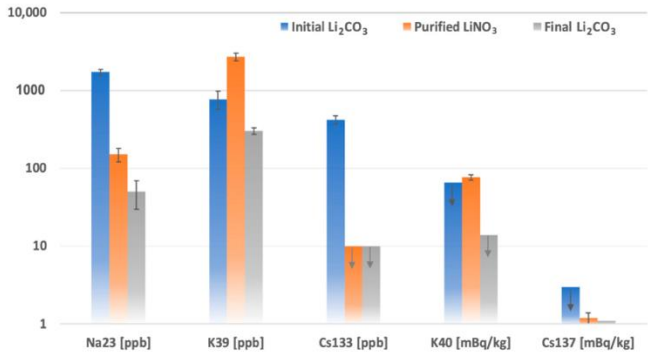
Unavailable

High Ra and Th

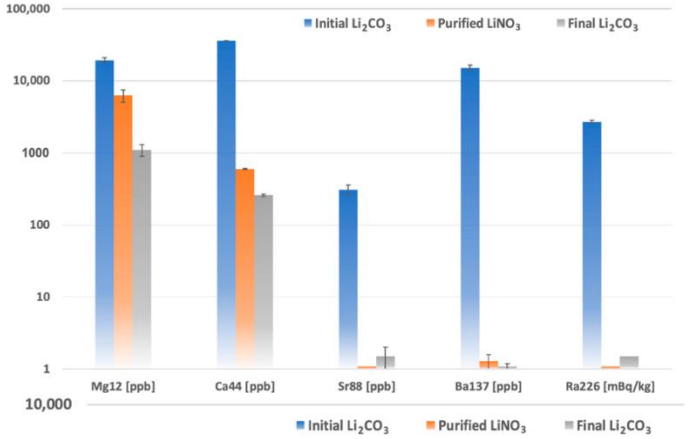
Unavailable

Impurities reduction in Li_2CO_3 with CUP purification

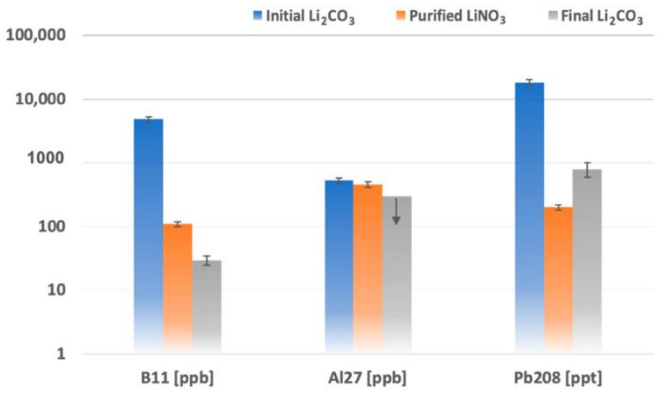
K/Cs



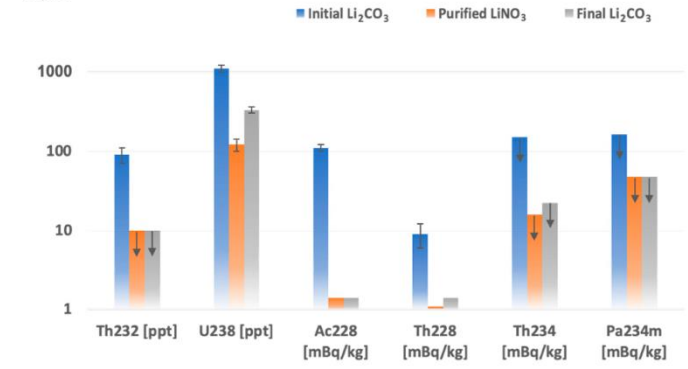
Ra/Ba



Pb



Th/U



Radioactivity reduction in Li_2CO_3 with CUP purification

Raw Li_2CO_3

Radionuclides	Activity [mBq/kg]	Purified $LiNO_3$	Final Li_2CO_3
^{40}K	≤ 66	77 ± 6	≤ 14
^{137}Cs	≤ 3	1.2 ± 0.2	≤ 1.0
^{234}Th	≤ 151	≤ 16	≤ 22
^{234m}Pa	≤ 162	≤ 47	≤ 47
^{226}Ra	2730 ± 140	≤ 1.0	≤ 1.5
^{228}Ac	110 ± 10	≤ 1.4	≤ 1.4
^{228}Th	9 ± 3	≤ 1.0	≤ 1.4

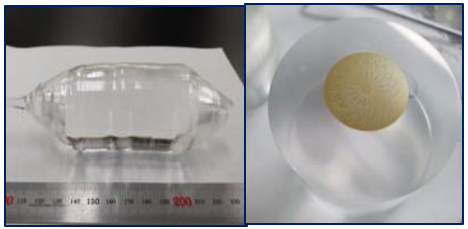
We have developed several methods of purification to remove K, Th, U, and Ra contamination

- For mass-scale purification (over 10 kg per month), the construction of a special purification facility will be required

Other candidates to be AMoRE-II detector

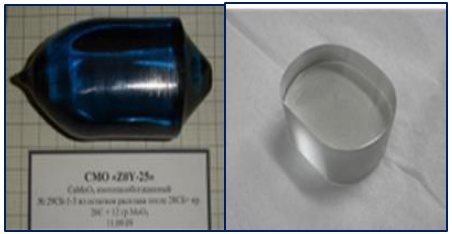
AMoRE-II crystal growing and purification R&D

Li_2CO_3 → Li_2MoO_4



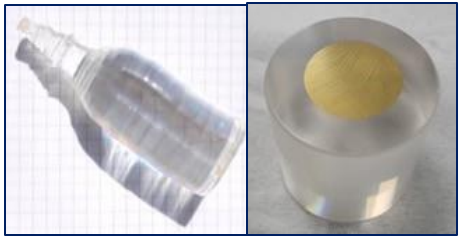
Best radiopurity achieved:
Frontiers of Physics 12(2024):1347162
Inorganics 11(10):410

$CaCO_3$ → $CaMoO_4$



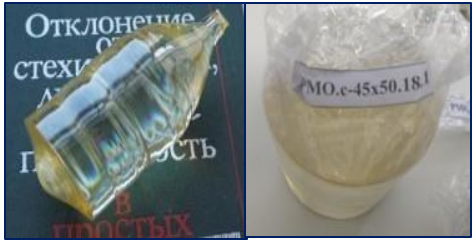
$^{48depl}Ca$ is required. Purification and recycling methods were developed at CUP:
Journal of Material Cycles and Waste Management 21(6)

Na_2CO_3 → $Na_2Mo_2O_7$



No commercially available pure enough product:
Inorganic Materials 56(8):867-874

Arch Pb → $PbMoO_4$



Archaeological Roman lead is required. Very difficult to recycle

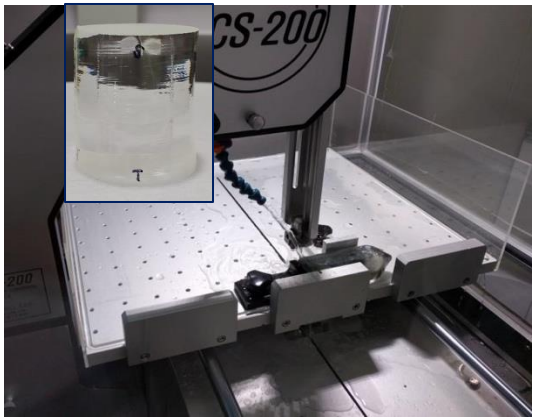
Crystal synthesis for AMoRE-II at CUP

Single-crystal production at CUP _ machining

Conventional Zcochralski pullers are used for the production of LMO crystals at CUP. The grown ingots are cut, polished, and rinsed at CUP to keep the surface clean.

Three conventional ZC pullers at CUP with Pt crucibles

Li_2MoO_4 ingot

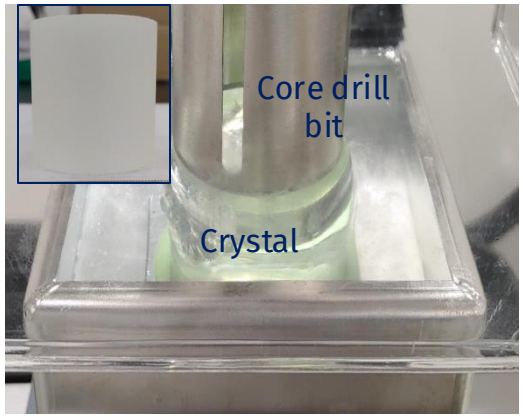


Cutting the shoulders out. The shoulders are re-used in the next growing circle.

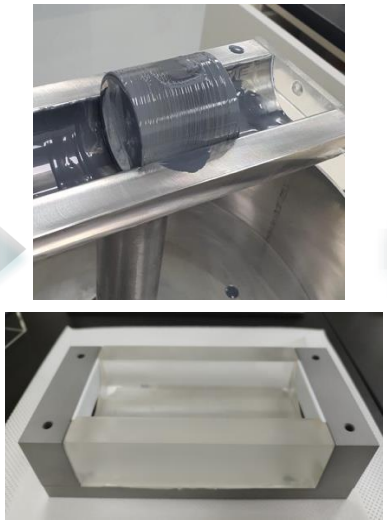
Crystal machining for AMoRE-II

Single-crystal production at CUP _ Surface treatment

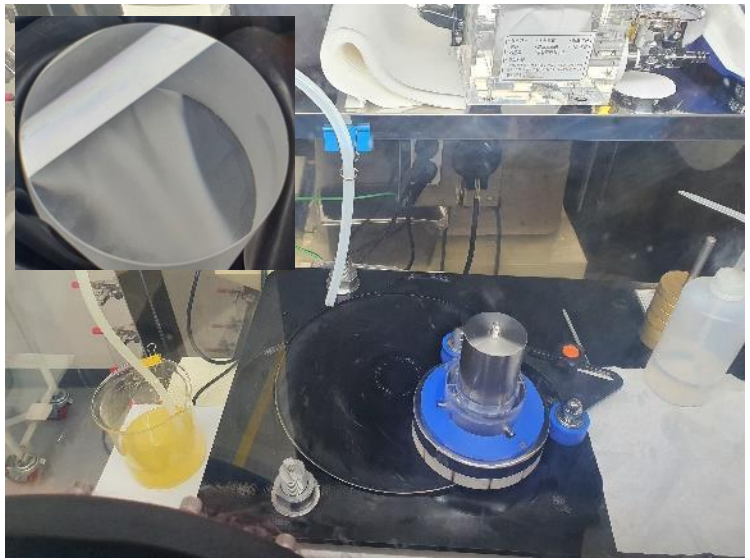
After the cutting and drilling, the crystals are lapped and polished in glovebox using high-purity oil and abrasives in pure N_2 environment to protect the crystal surface from damage and contamination. LMO crystals are highly hygroscopic. In short, the surface of machined crystals are degreased and cleaned.



Coring



Side lapping: SUS jig or Quartz jig

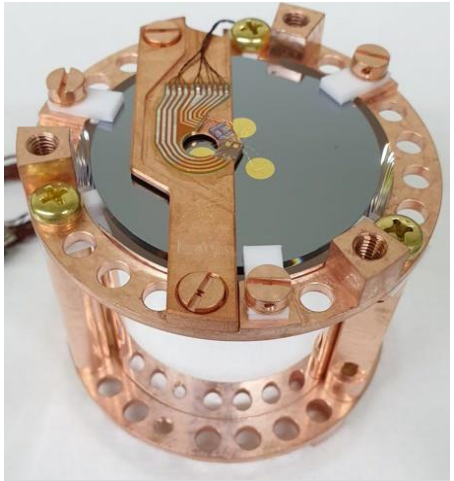


Flat side polishing

Purity control of AMoRE-II crystals with ICP-MS

- A segregation factor of over five orders of magnitude could be achieved with LMO crystal growing at CUP with Conventional CZ [JINST 15(07):C07035-C07035].
- All enriched AMoRE-II crystals are tested at CUP with ICP-MS and show the purity as shown in the table.
- Final radiopurity can be derived after the completion of full detector assembly.
- AMoRE-I detector se up showed purity of LMO crystals at the level of a few μBq/kg.

Sample ID		K	Ba	Sr	Zr	Ir	Pb	Th	U
		(ppb)	(ppb)	ppt	ppt	ppt	ppt	ppt	ppt
EnrLMO crystal	Top	<40	<3	<50	<100	<50	<200	<6	<6
	Bottom	<40	<3	<50	<100	<50	<200	<6	<6



AMoRE-II crystal assembled in copper holder

Towards COSINE-200

01 Raw material purification

- Based on recrystallization from water.
- 70 kg/month is production efficiency for one crystallization run.

02 Single-crystal growing

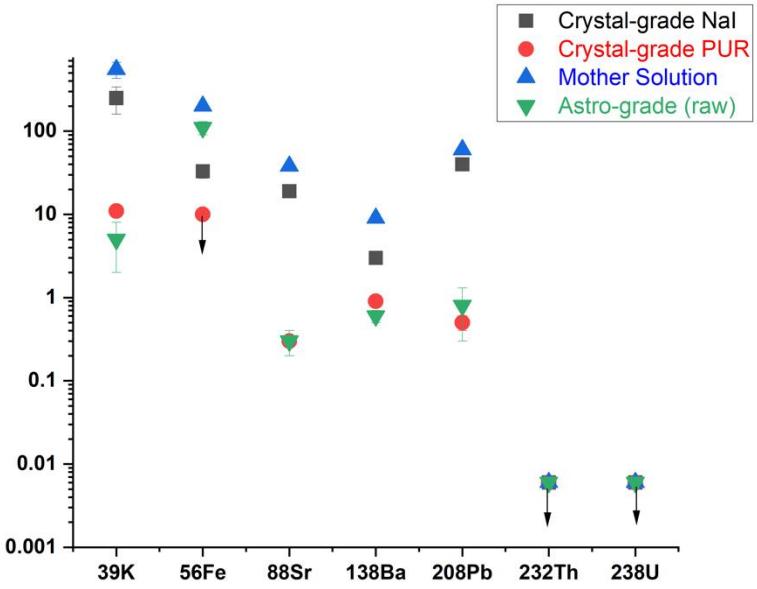
- Kyropoulos growers with a quartz crucible.
- Small-size grower was used as a proof of principles.
- Full-size crystal growing is under investigation.

Mass-purification facility

Small-size grower
1 kg ingot

Full-size crystal grower
20 kg ingot

Mass-purification of commercial Crystal Grade NaI powder



Impurities content in purified (PUR) and raw NaI materials

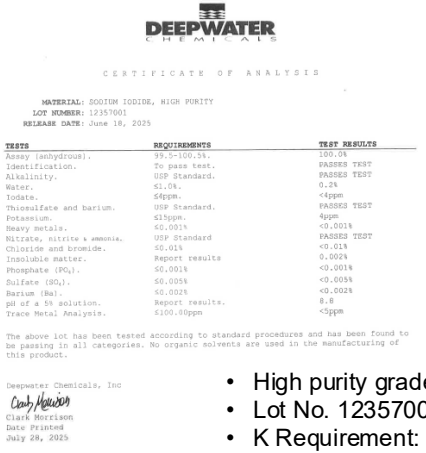
03

Impurities reduction with CUP purification process

- Crystal-grade (CG) NaI powder is used as a starting material for purification, costing a few hundred USD/kg.
- Astro-grade (AG) powder is commercially available and suitable for COSINE-200 crystal growing, costing a few thousand USD/kg.
- In one crystallization run, we achieved AG purity, and the mother solution is recyclable.
- CUP-grade purified powder is entirely fit for the COSINE-200 applications.

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

NaI raw powder from Deep Water Co.



DEEPWATER CHEMICALS

CERTIFICATE OF ANALYSIS

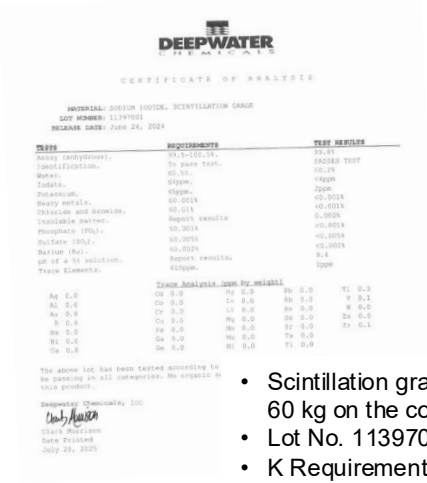
MATERIAL: SODIUM IODIDE, HIGH PURITY
LOT NUMBER: 12357001
RELEASE DATE: June 18, 2025

TESTS	REQUIREMENTS	TEST RESULTS
Assay (anhydrous).	99.5-100.5%.	100.0%
Identification.	To pass test.	PASSES TEST
Alkalinity.	USP Standard.	PASSES TEST
Water.	≤1.0%.	0.2%
Iodate.	≤4ppm.	<4ppm
Thiosulfate and barium.	USP Standard.	PASSES TEST
Potassium.	≤15ppm.	4ppm
Heavy metals.	≤0.001%	<0.001%
Nitrate, nitrite & ammonia.	USP Standard	PASSES TEST
Chloride and bromide.	≤0.01%	<0.01%
Insoluble matter.	Report results	0.002%
Phosphate (PO_4).	≤0.001%	<0.001%
Sulfate (SO_4).	≤0.005%	<0.005%
Barium (Ba).	≤0.002%	<0.002%
pH of a 5% solution.	Report results.	8.8
Trace Metal Analysis.	≤100.00ppm	<5ppm

The above lot has been tested according to standard procedures and has been found to be passing in all categories. No organic solvents are used in the manufacturing of this product.

Deepwater Chemicals, Inc
Clark Morrison
Clark Morrison
Date Printed
July 28, 2025

- High purity grade – 20 kg in stuck
- Lot No. 12357001
- K Requirement: ≤15ppm
- **Test results: 4 ppm**



DEEPWATER CHEMICALS

CERTIFICATE OF ANALYSIS

MATERIAL: SODIUM IODIDE, SCINTILLATION GRADE
LOT NUMBER: 11397001
RELEASE DATE: June 24, 2024

TESTS	REQUIREMENTS	TEST RESULTS
Assay (anhydrous).	99.5-100.5%.	99.6%
Identification.	To pass test.	PASSES TEST
Water.	≤0.5%.	<0.2%
Iodate.	≤4ppm.	<4ppm
Potassium.	≤5ppm.	2ppm
Heavy metals.	≤0.001%	<0.001%
Chloride and bromide.	≤0.01%	<0.001%
Insoluble matter.	Report results	0.002%
Phosphate (PO_4).	≤0.001%	<0.001%
Sulfate (SO_4).	≤0.005%	<0.005%
Barium (Ba).	≤0.002%	<0.002%
pH of a 5% solution.	Report results.	9.4
Trace Elements.	≤100ppm.	1ppm

Trace Analysis (ppm By weight)

Ag	0.0	Cd	0.0	Mg	0.0	Pb	0.0	Tl	0.3
Al	0.0	Co	0.0	In	0.0	Rb	0.0	V	0.1
As	0.0	Cr	0.0	Li	0.0	Sn	0.0	W	0.0
B	0.0	Cu	0.0	Mg	0.0	Sr	0.0	Zn	0.0
Ba	0.0	Fe	0.0	Mn	0.0	Ti	0.0	Zr	0.1
Bi	0.0	Ga	0.0	Mo	0.0	Ta	0.0		
Ca	0.0	Ge	0.0	Ni	0.0	Ti	0.0		

The above lot has been tested according to standard procedures and has been found to be passing in all categories. No organic solvents are used in the manufacturing of this product.

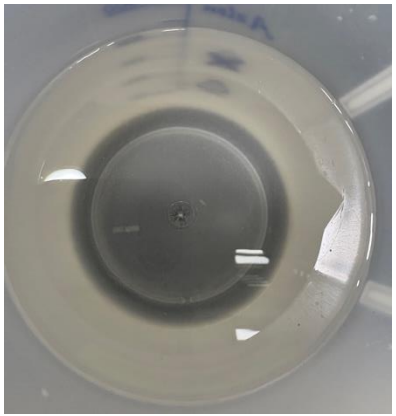
Deepwater Chemicals, Inc
Clark Morrison
Clark Morrison
Date Printed
July 28, 2025

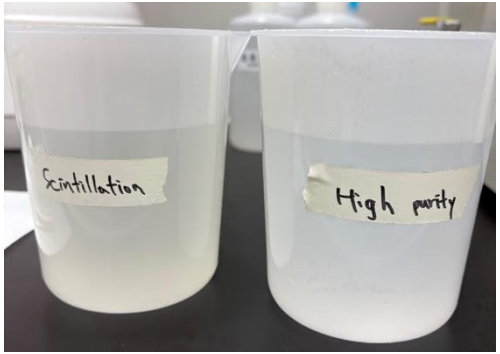
- Scintillation grade – 40 kg in stuck, 60 kg on the contract.
- Lot No. 11397001
- K Requirement: ≤5ppm
- **Test results: 2 ppm**

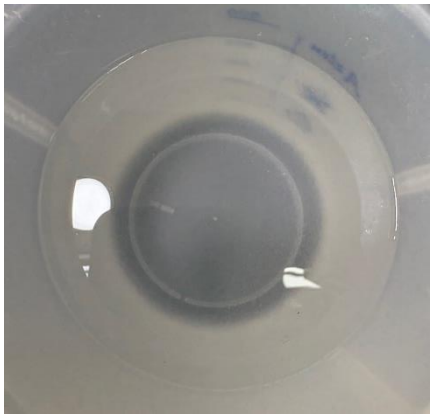
Sample	Al	K	Sr	Ba	Pb	Th	U
	ppb					ppt	
High purity grade	<100	2,406	63.6	240.1	1.4	<4	24
Scintillation grade	**<100**	**339**	**68.7**	**109.7**	**2.1**	**<4**	**19**
Merck, 2020, TP1224159	110	248	19	2.9	40	<6	<6
Merck, 2021, TP1356959	2.7	730	42	146	58	<100	415

NaI raw powder from Deep Water Co.

◆ **Dissolving Test**







Scintillation grade

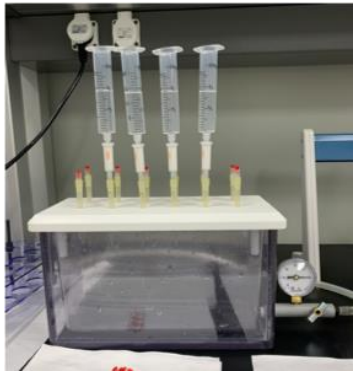
High purity grade

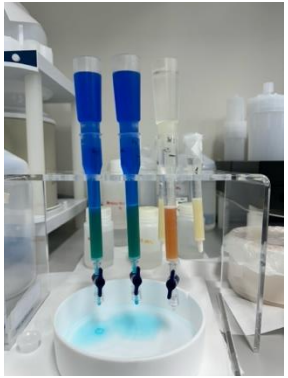
- Both powders had a few insoluble particles, but overall, they were clean.
- The scintillation grade powder has a darker color, brownish and grayish.

ICP-MS and chemical extraction facilities at CUP

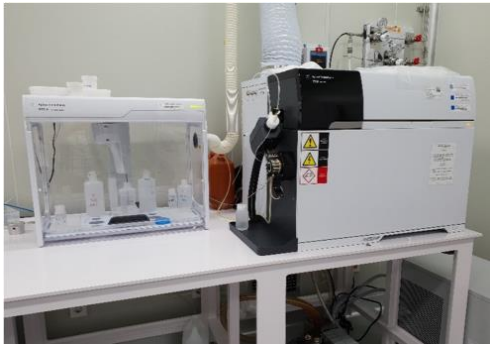
Class 1000 clean room

Microwave ashing





Extractive chromatography



ICP-MS Agilent 7900

Microwave digestion

ICP-MS Detection limits

- Copper and Brass: 0.26 ppt for Th and 0.1 ppt for U
- Vikuiti polymer film: 3 ppt for Th and U
- HDPE: 1.2 ppt for Th and U
- MoO_3 powder: 2.3 ppt for Th and 3.3 ppt for U

* Appl. Radiat. and Isot. (2023),110673

Cu holder manufacturing from the original pure NOSV bulk

01 Selection of manufacturer

Posts (unit of the copper frame for Amore-II)

	Th, ppt	U, ppt
NOSV 2021	0.26 ± 0.01	0.29 ± 0.06
After 20 µm surface removal		
Company 1 ES JeongMill	<0.5	0.43 ± 0.09
Company 2 Taeseong Tech	<0.5	0.60 ± 0.09
Company 3 ShinHan TC	77.3 ± 1.8	12.2 ± 0.4

02 Where is the source of contamination when machining?

	Mill	Thread	NOSV-Cu 2021 bulk
Surface thickness removed, µm	~1	~4	
Th, pg/g of removed surface	80 ± 10	1370 ± 150	0.26 ± 0.01
U, pg/g of removed surface	30 ± 5	300 ± 30	0.29 ± 0.06

Cu screws manufacturing from the riginal OFE bulk

- The M4 screws were cleaned step-wise and effectiveness of each step was checked individually.
- The screws after surface cleaning were dissolved entirely, treated with UTEVA-SPE and measured with ICP-MS

Raw material	Th, pg/g of Cu	U, pg/g of Cu
OFE Aurubis 2021	0.98 ± 0.14	0.83 ± 0.11

1.No cleaning

2. Kerosene + Ethanol

3. Oxalic acid

4. Nitric acid

Th, pg/g of screw	27 ± 3	30 ± 3	14 ± 2	11 ± 2
U, pg/g of screw	8 ± 2	8 ± 2	3 ± 1	2 ± 0.5

Screws_Brass_Sunco_replace the OFE-Cu 2021 screws

- Screws made of OFE-Cu 2021 were found unacceptably contaminated, we tested brass screws made by Sunco comp., Japan.
- All brass screws were degreased and etched with HNO_3.
- The brass crews were cleaned well and can be used in AMoRE-II detector assembly.

Raw material, full-body meas.	Th, pg/g of	U, pg/g of Cu
2. No cleaning brass	89.25 ± 0.39	20.75 ± 0.56
3. Sonication with ethanol	5.69 ± 0.14	1.39 ± 0.15
4. HNO_3 etching	1.43 ± 0.13	0.49 ± 0.12

Testing 1st batch for 90 crystal holders

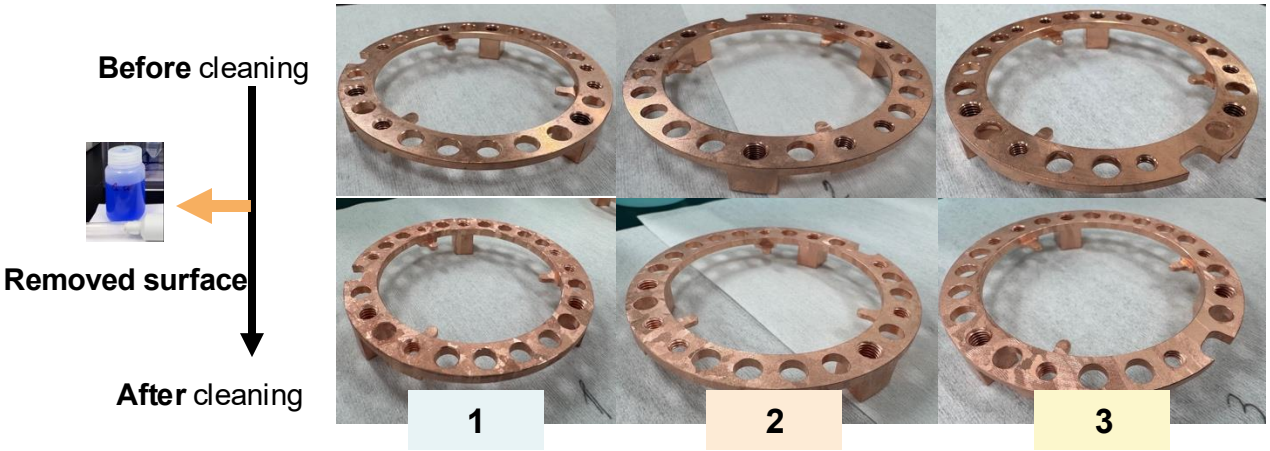
2024	M4-4	M4-6	M5-12	M5-15
Th, pg/g	0.67 ± 0.11	1.16 ± 0.13	1.03 ± 0.13	0.68 ± 0.08
U, pg/g	0.17 ± 0.04	0.48 ± 0.05	0.34 ± 0.03	0.22 ± 0.06

Testing 2nd batch for entire 270 crystal holders

2025	M4-4	M4-6	M5-12	M5-15
Th, pg/g	<0.67	<0.67	<0.67	<0.67
U, pg/g	0.15 ± 0.04	0.16 ± 0.04	0.16 ± 0.03	0.09 ± 0.03

Top and bottom circle plates_randomly contaminated

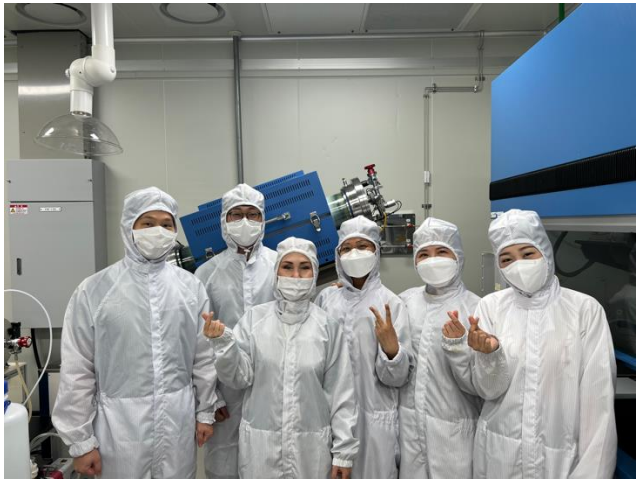
- Randomly selected samples were cleaned and measured individually.
- The surface etch only was measured, but the entire body was preserved for further use.



2025 results	1			2			3			NOSV bulk
Thickness, µm	2-4	4-8	**8-12**	2-5	5-10	**10-15**	2-5	5-10	**10-15**	-
Th, pg/g of dissolved surface	521.4	63.5	**12.8**	50.6	6.0	**1.6**	19.6	8.0	**0.32**	0.26 ± 0.01
U, pg/g of dissolved surface	123.2	15.5	**5.4**	20.4	6.9	**2.5**	22.0	4.0	**8.2**	0.29 ± 0.06

Summary

- The chemistry purification labs have been established and are successfully running at CUP.
- Various purification methods were developed for the purification and mass production of low-radioactive materials for CUP projects.
- The purity of the produced materials is confirmed with ICP-MS and HPGe at CUP.
- Surface machining and cleaning of copper, lead, aluminum, etc.; bulk contamination studies; etc., are ongoing.



Thanks for coming!