CENTER FOR UNDERGROUND PHYSICS

Th and U assay in high-purity Cu @ CUP

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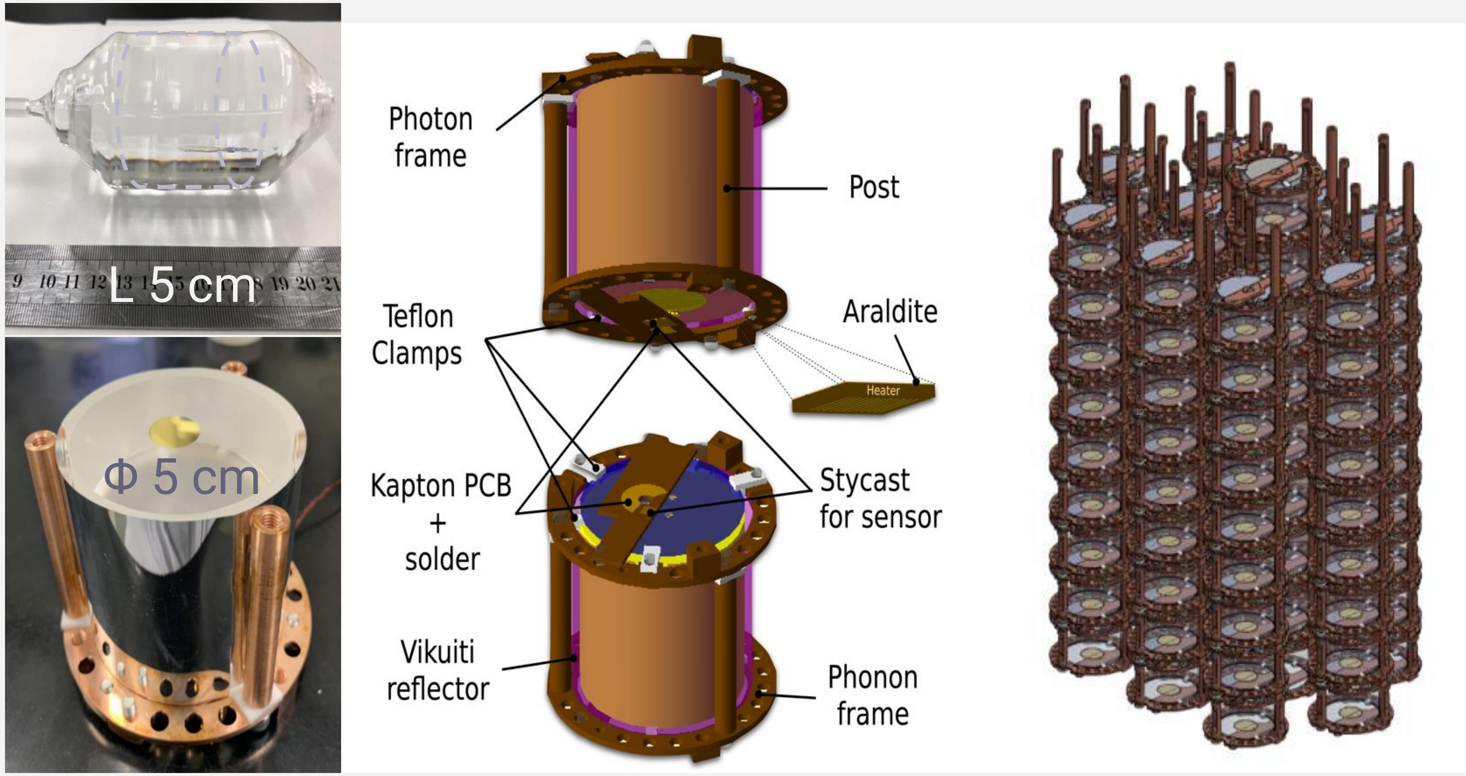
Motivation:



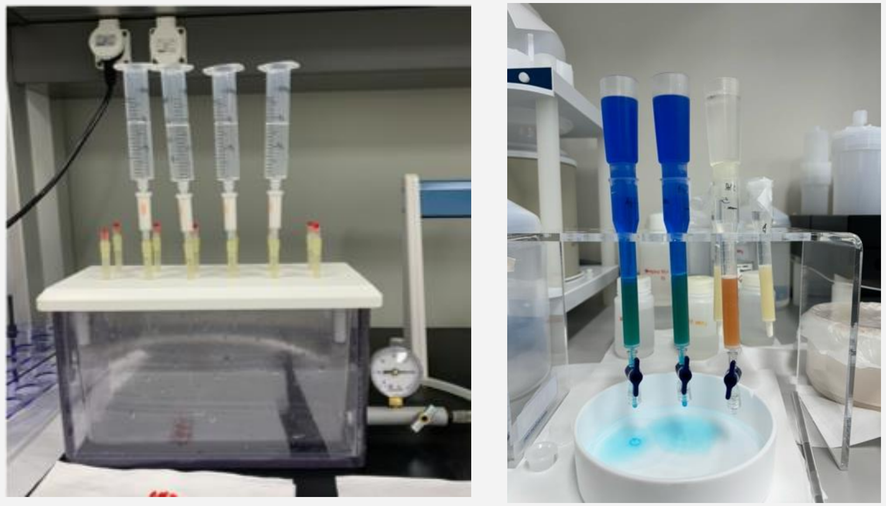
I. CUP-grown $Li_2{}^{100}MoO_4$ crystal ingot; II. $Li_2{}^{100}MoO_4$ scintillation element after cutting, polishing, surface cleaning, and gold film deposition procedures; III. **Copper crystal holder frame** and near-crystal components; IV. Detector tower consisting of 360 enriched $Li_2{}^{100}MoO_4$ crystals.

- AMoRE searches for the neutrinoless double beta decay (*0νββ*) of ^{100}Mo using 360 bolometric $Li_2{}^{100}MoO_4$ crystals.
- *Located in Yemi Underground Lab ~1000 meters deep in Jeongseon, Korea.*
- Background index of ~10^{-4} ckky *with masses of the studied isotope of interest of 100 kg [arXiv:1512.05957v1].*
- *The development and involvement of radiochemical purification and separation methods are essential for the background control [Appl Radiat Isot 194(216):110673 (2023); Front. in Phys. 1362209 (2024)].*
- *The construction of the detector itself, its peripheral sub-systems and shielding needs* the development of special surface cleaning procedures and highly sensitive radio-assay methods [Eur. Phys.J. C 85:9 (2025)].

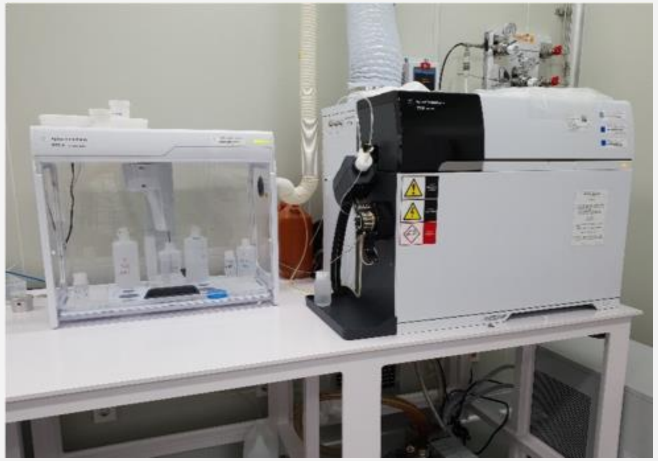
ICP-MS and chemical extraction facilities at CUP

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

SPE-ICPMS procedure for Th and U analysis in Cu at CUP

1. pre-washing

- 60 - 80 mL 0.1M HCl by 20 mL additions
- Validation: 15 mL 0.1M HCl

2. Activation

- 5 mL 5M HNO_3

3. Sample loading

- 20 -25 mL 5M HNO_3 matrix sample

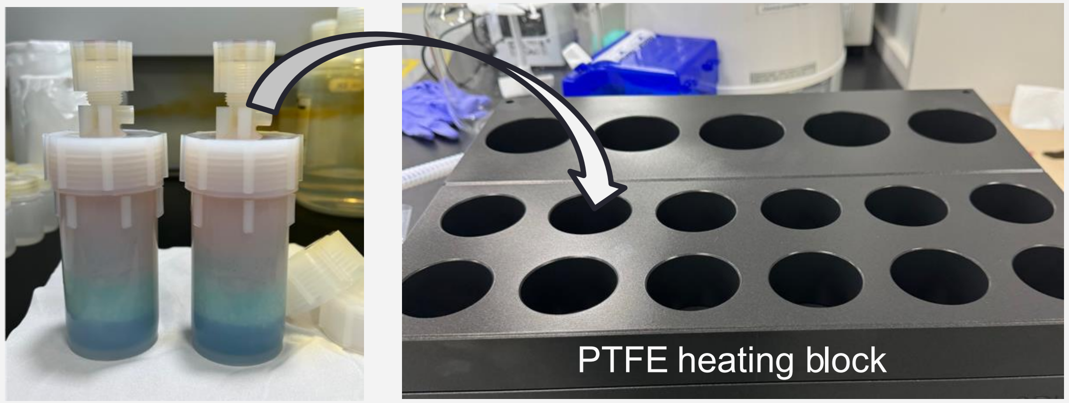
4. Washing

- 4 mL 5M HNO_3 by 2 mL additions

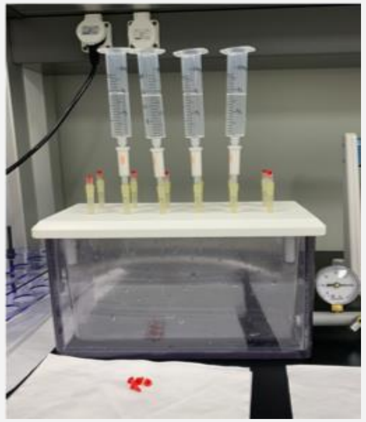
5. Elution

- 10 mL 0.1M HCl

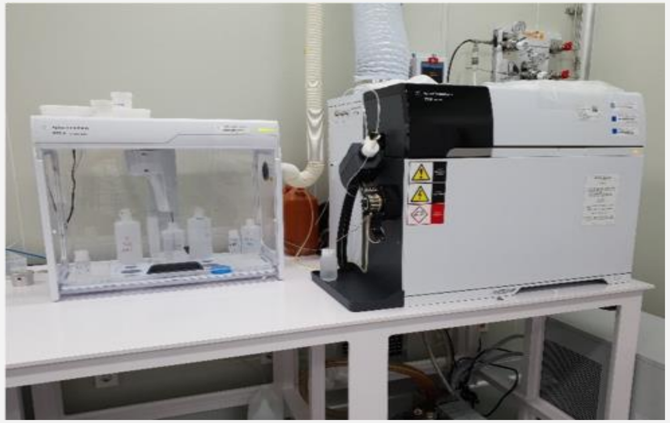
1. Sample decomposition



2. UTEVA SPE in vacuum box

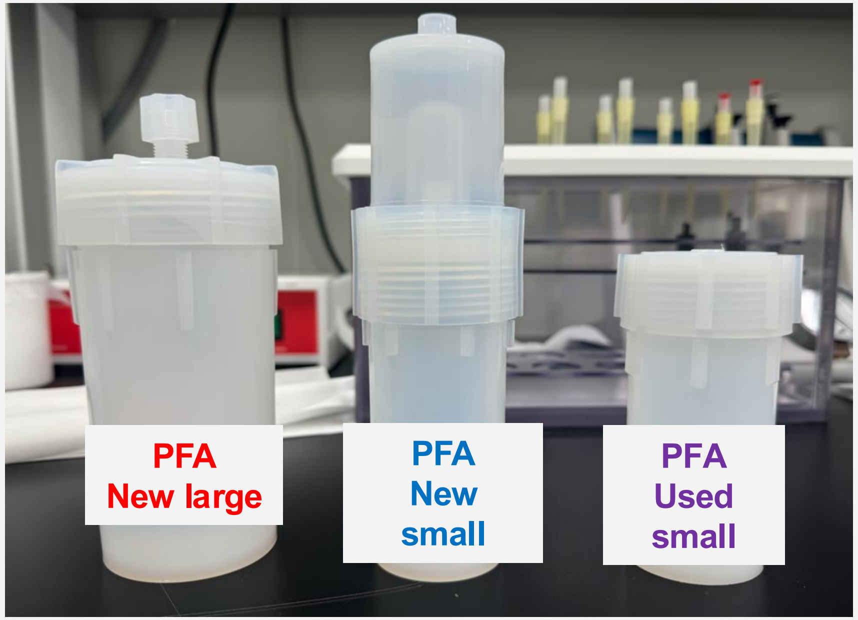


3. ICP-MS detection at CUP with Agilent 7900



Blank and recovery check in every run

- ✓ Optimal labware is selected and used for blank and recovery checks. For the recovery, we always do the spike sample.
- ✓ Measured with UTEVA-SPE
- ✓ Results shown for 10 mL of elution



	Th, pg/g of eluate	U, pg/g of eluate
PFA digestion vessel New large	0.015 – 0.020	0.010 - 0.015
PFA digestion vessel New small	0.010 - 0.020	0.010 - 0.020
PFA digestion vessel Old small	0.090 - 0.250	0.040 - 0.150
Quartz	0.080 – 0.120	0.030 – 0.050
PP beaker used	0.150 – 0.850	0.100 – 0.200
PP beaker new	0.100 – 0.250	0.010 – 0.060
PTFE beaker	0.100 – 0.450	0.050 – 0.150

- ✓ To maximize the analysis precision, the blank from each vessel used is prepared before the sample treatment.

Th and U analysis in Bulk Cu at CUP

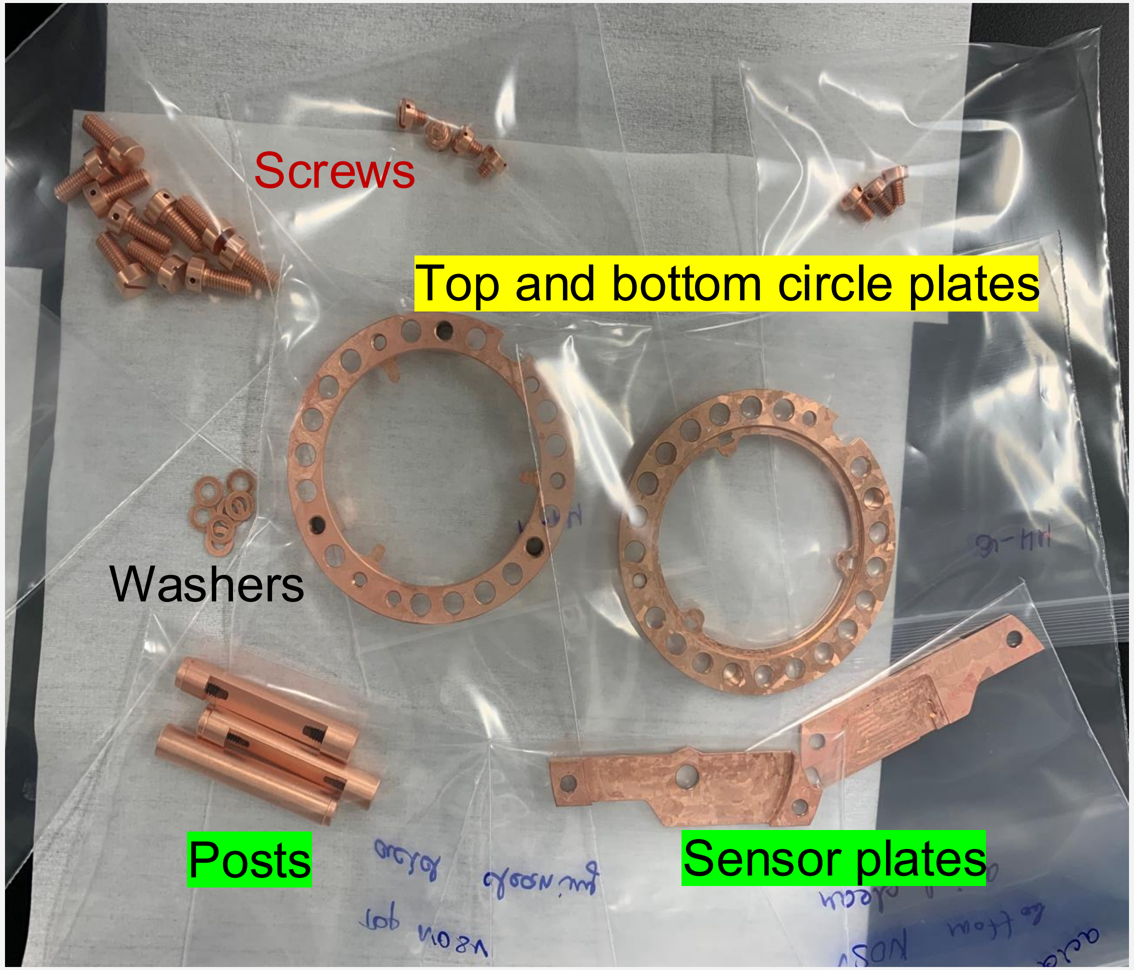
Procedural detection limits based on blank measurements (PFA digestion vessels):

✓ ***0.1 pg/g of Cu for U*** ✓ ***0.12 pg/g of Cu for Th***

	^{232}Th, pg/g of Cu	^{238}U, pg/g of Cu	Notes
NOSV 2014 (October, 2019 meas.)	**4.3 ± 0.29**	**1.66 ± 0.04**	**CUP, SPE-ICPMS**
NOSV 2014 (May, 2019 meas.)	**5.13 ± 1.2**	**1.55 ± 0.7**	**CUP, SPE-ICPMS**
NOSV Aurubis 2016	**0.34 ± 0.12**	**0.29 ± 0.04**	**CUP, SPE-ICPMS**
NOSV Aurubis 2021	**0.26 ± 0.01**	**0.29 ± 0.06**	**CUP, SPE-ICPMS**
Aurubis Cu C10100	0.46 ± 0.06	0.21 ± 0.06	MAJORANA, SPE-ICPMS
Aurubis, NOSV Cu	<3.2	-	CUORE, SPE-NAA
Aurubis, NOSV Cu	<0.5	<5.3	CUORE, NAA+HPGe
OFE Mitsubishi	**2.19 ± 0.21**	**1.28 ±0.19**	**CUP, SPE-ICPMS**
Mitsubishi Cu C10100	2.12 ± 0.39	2.25 ± 0.15	MAJORANA, SPE-ICPMS
OFE Aurubis 2018	**0.98 ± 0.08**	**1.01 ± 0.05**	**CUP, SPE-ICPMS**
OFE Aurubis 2021	**0.98 ± 0.14**	**0.83 ± 0.11**	**CUP, SPE-ICPMS**
OFE Aurubis	<15.7	<4.4	CUORE, NAA+HPGe
OFE Wonil 2023 (Korea)	**0.75 ± 0.15**	**0.43 ± 0.12**	**CUP, SPE-ICPMS**

NOSV Aurubis 2021 was selected for AMoRE-II detector holders

Crystal holder units for AMoRE-II Phase 1 (90 crystals out of 360)



1. **Screws** were initially designed to be machined from OFE-Cu due to its higher mechanical strength. However, due to high contamination caused by the machined surface, we replaced them with commercial brass screws.
2. **Sensor plates** were the easiest to machine and clean.
3. **Posts** were tough to clean, have threaded hollows hidden from sonication. The exterior surface was contaminated up to 30 μm depth.
4. **Top and bottom circle plates** have threaded areas, which can introduce additional contamination.

Cu-OFE 2021 screws _ unacceptable purity

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

1.No cleaning

2. Kerosene + Ethanol

3. Oxalic acid

4. Nitric acid

	1	2	3	4
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

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

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**

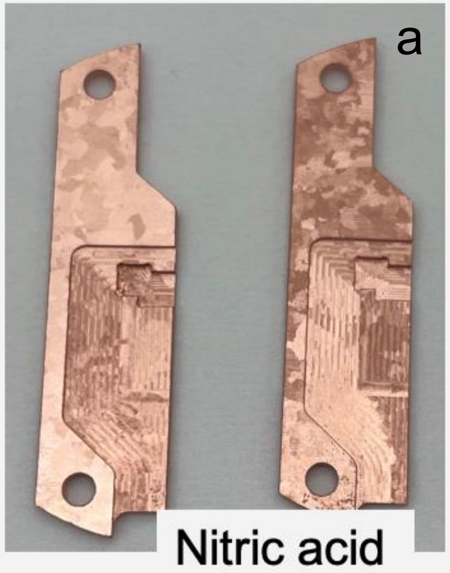
Testing 1st batch for 90 crystal holders

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

Sensor plates after HNO_3 etching are Ok!



- Sensor plates were degreased, etched in HNO_3 under sonication to remove ~ 10 μm surface, passivated, and the entire bodies were measured.
- The mill surface (flat, simple) is less damaged and contaminated than lathe surface (round) and tapping (screw or thread), so surface is cleaned easier.
- Each machined batch was checked on purity.
- Based on simulation results, these levels are suitable for AMoRE-II.

	Th		U, ppt	
	pg/g of sensor plate	pg/sensor plate	pg/g of sensor plate	pg/sensor plate
NOSV-Cu bulk 2021	**0.26 ± 0.01**	**3.3 ± 0.1**	**0.29 ± 0.06**	**3.7** ± 0.8
Sample a-1 - 2023	0.34 ± 0.09	4.3 ± 1.1	0.50 ± 0.05	6.4 ± 0.6
Sample a-2 - 2023	0.33 ± 0.12	4.2 ± 1.2	0.41 ± 0.08	5.2 ± 1.0
Sample b-1 – 2024	0.54 ± 0.07	5.1 ± 0.7	0.35 ± 0.12	3.3 ± 1.1
Sample b-2 - 2024	0.24 ± 0.15	2.3 ± 1.4	0.38 ± 0.21	3.61 ± 2.0

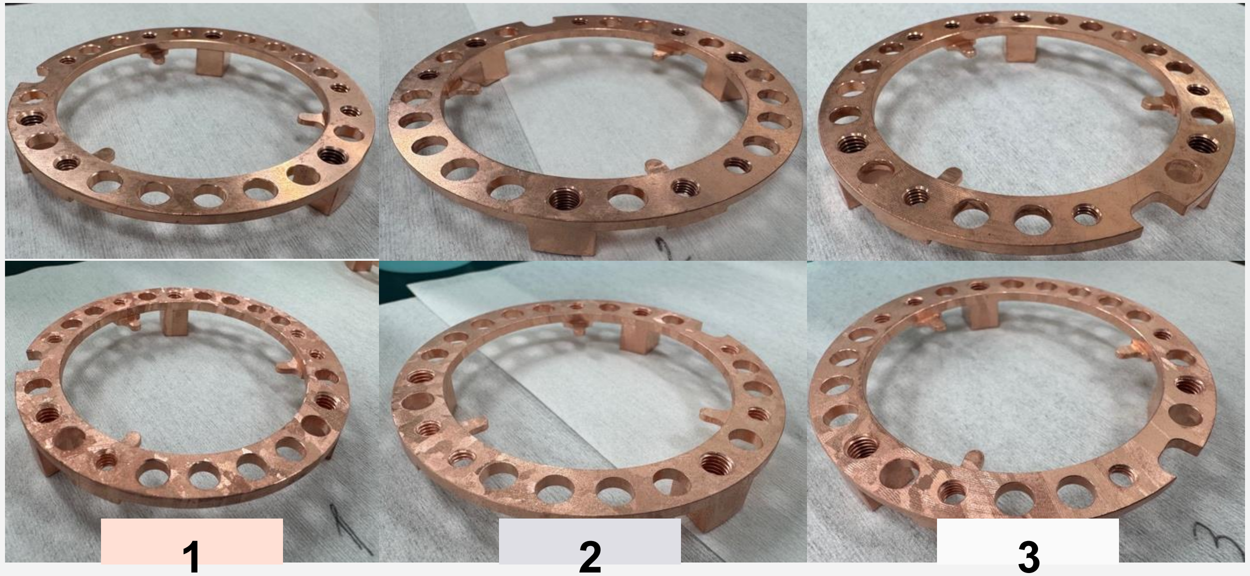
Top and bottom circle plates_randomly contaminated

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

Before cleaning

Removed surface

After cleaning



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

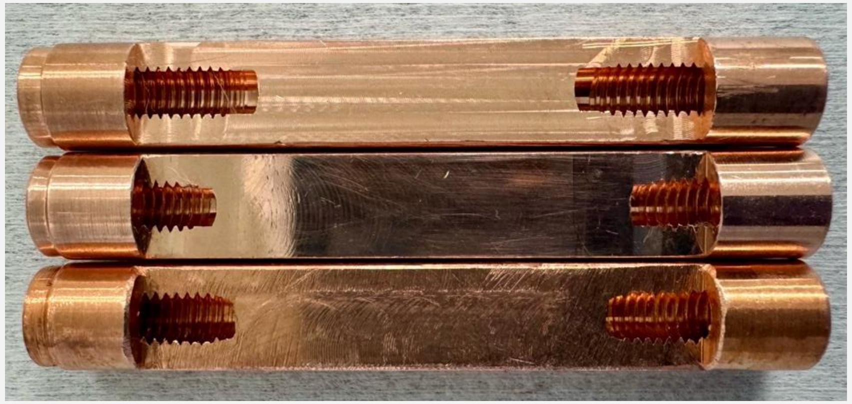
Top and bottom circle plates_30 µm to be removed

- We continued with **sample 1**, the dirtier one, to remove the surface and measure it at a deeper level.
- The contamination for Th and U did not change significantly in the range of 8-20 µm surface thickness.
- For the 21-26 µm surface thickness, the contamination reduced to the acceptable level.
- Conclusion → we must remove ~30 µm of the surface from each top and bottom circle.

	Sample 1 (~45 g)					NOSV bulk
Thickness removed, µm	2-4	4-8	**8-12**	**13-20**	**21-26**	-
Th, pg/g of dissolved surface	521.4	63.5	**12.8**	**12.2**	**5.4**	0.26 ± 0.01
U, pg/g of dissolved surface	123.2	15.5	**5.4**	**6.9**	**3.4**	0.29 ± 0.06

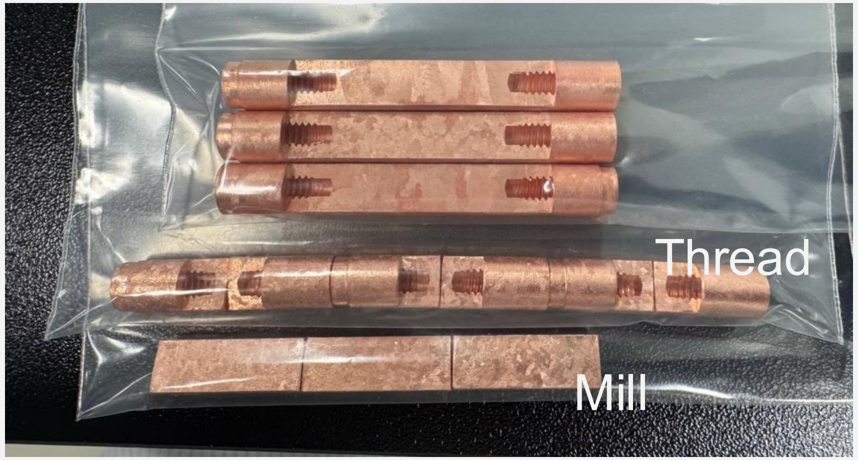
Posts were the biggest problem but solved!

Selection of the manufacturer



	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

Where is the source of contamination?



	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**

Summary - Highlights

- With SPE-ICPMS at CUP, we selected suitable AMoRE-II copper to prepare crystal holders.
- All units of the crystal holder were checked for purity after the machining.
- Cu-OFE screws were found to be highly contaminated and replaced with commercially available brass screws.
- A dedicated company implemented additional precautions to prepare the posts with minimal and removable surface contamination.

Thanks for your attention!