CENTER FOR UNDERGROUND PHYSICS

Preparation of the precursors for AMoRE-II crystals synthesis

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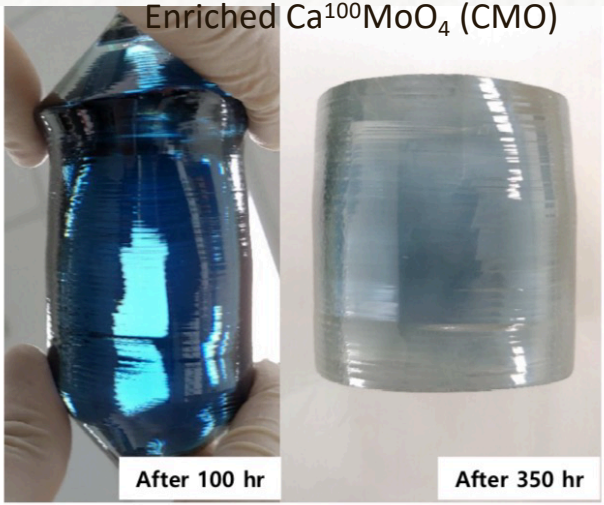
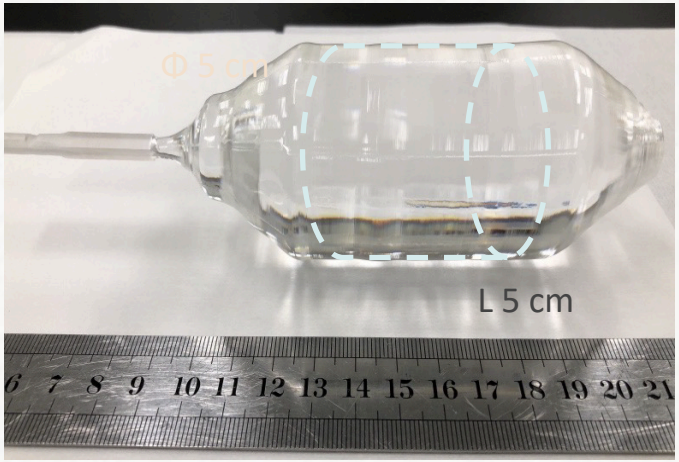
AMoRE experiment

The Advanced Molybdenum based Rare process Experiment is a series of experiments searching for the neutrinoless double beta ($0\nu\beta\beta$) decay of the ^{100}Mo isotope with cryogenic detectors using molybdate ($^{100}MoO_4$)-based bolometric crystal detector. Various molybdate crystals such as ^{48}dep$Ca^{100}MoO_4$, $Li_2{}^{100}MoO_4$, $Na_2Mo_2O_7$, and $PbMoO_4$ for AMoRE-II, the second phase of the experiments, have grown and tested for their performances including background radiation levels.

Enriched $Ca^{100}MoO_4$ (CMO)



Enriched $Li_2{}^{100}MoO_4$ (LMO)

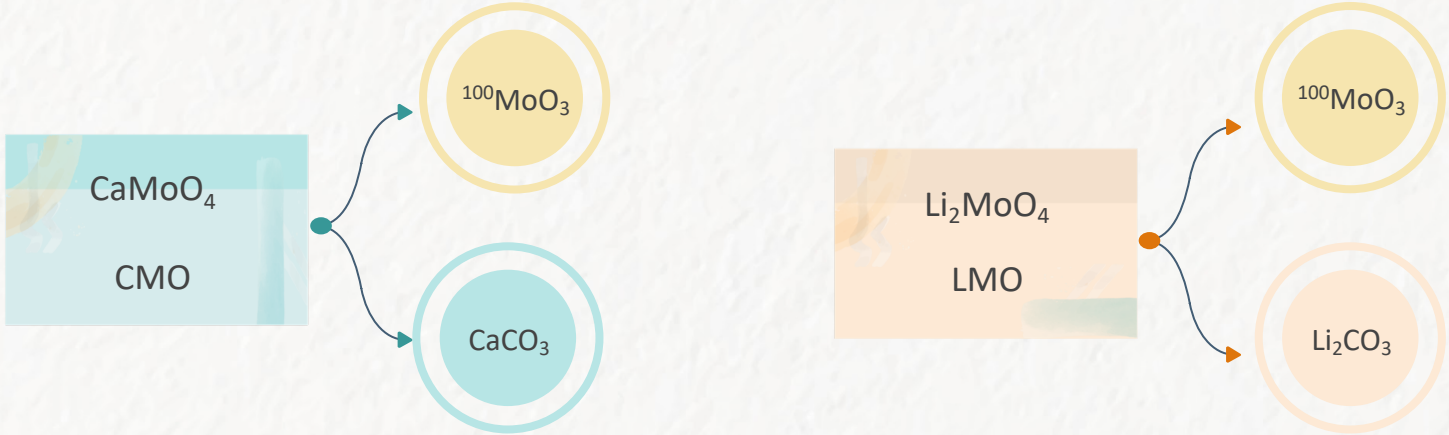


Background requirements for AMoRE

The process requires that the detector apparatus and its components including the scintillating crystals and thus the initial materials used for crystal growth be extremely low in radioactive isotopes having decays, which may generate background noise signals in the ROI.

	AMoRE-I	AMoRE-II
	$<10^{-3}$ ckky in ROI ~ 3 kg ^{100}Mo 18 crystals (5 LMO + 13 CMO)	$<10^{-4}$ ckky in ROI ~ 100 kg ^{100}Mo ~ 420 crystals
Contribution to background from Th and U chains, ppt		
Internal crystal	<100	<10

Precursors for the crystals production



Assuming a segregation of the impurities in the melt after the crystal growing, the concentration of the contaminants in the raw powders is expected to be less than 100 ppt

Outline

01 $^{100}MoO_3$
Purification, recovery and recycling

02 Li_2CO_3
Initial material selection, purification

03 $CaCO_3$
Purification, recovery and recycling

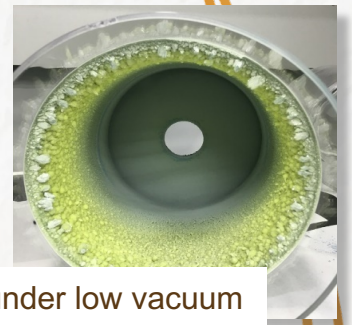
01 $^{100}MoO_3$

Purification, recovery and recycling

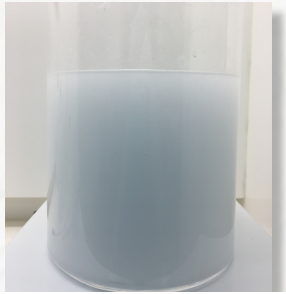
Enriched molybdenum oxide purification, mass-production and analysis

5 kg/month purification capacity at CUP

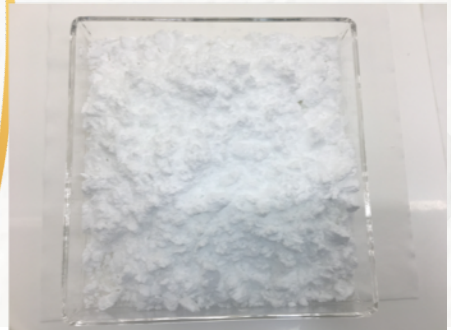
~99% recovery efficiency for the process

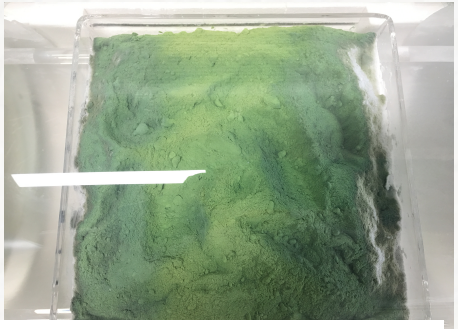


1. sublimation under low vacuum



2. dissolving, co-precipitation, filtering





3. synthesis of ammonium polymolybdate powder and its annealing

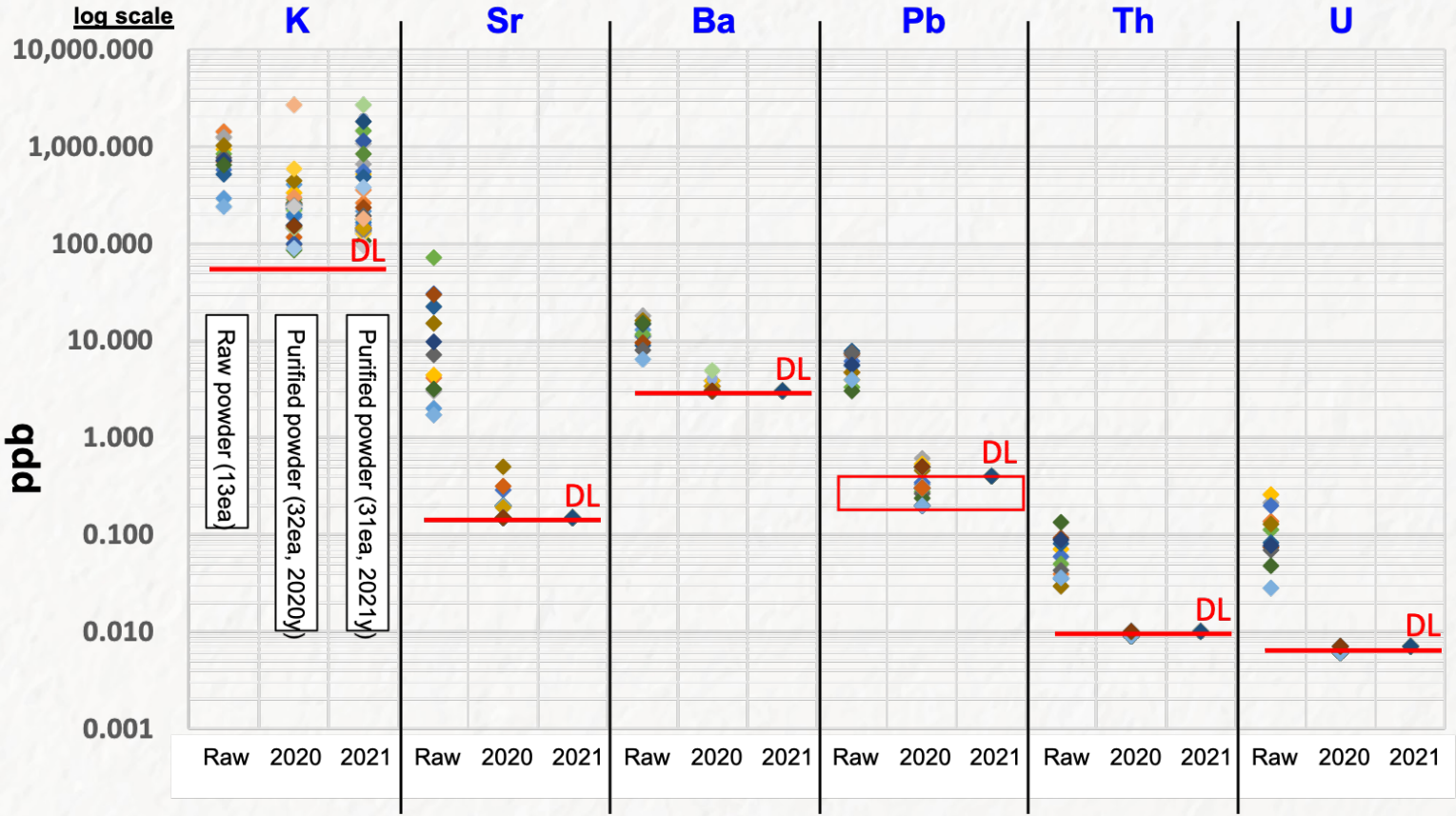
4. final $^{100}MoO_3$ powder collection and its storage at Y2L A5

Analysis of purified $^{100}MoO_3$ powders with ICP-MS at CUP

experiment #	Al, ppb	K, ppb	Cr, ppb	Mn, ppb	Fe, ppb	Ni, ppb	Cu, ppb	W, ppb	Sr, ppb	Ba, ppb	Pb, ppb	Th, ppt	U, ppt
Raw 100MoO3 powder (lot #3497)	1399	938	<300	<30	504	1073	<200	670	4	11	4.0	70	257
Purified products (examples)													
1	585	409	<200	<30	39	<20	<200	33	<0.15	3.9	0.33	<10	<7
11	630	253	<400	<30	26	<20	<200	43	<0.20	<3.0	<0.20	<9	<6
21	<30	246	<200	<30	33	<20	<200	38	<0.15	<3.0	<0.5	<10	<7
31	<30	150	<200	<30	26	<20	<200	648	<0.15	<3.0	<0.5	<10	<7
51	<100	146	<200	<30	18	<20	<200	601	<0.15	<3.0	<0.4	<10	<7

- $^{100}MoO_3$ powder is dissolved in a strong HCl using microwave digestion system Ethos Easy, Milestone.
- 1 wt% solution is analyzed with ICP-MS using He and UHMI modes.
- Standard - addition calibration to minimize matrix effect

Reduction of impurities in the $^{100}MoO_3$ produced



HPGe array measurement of purified $^{100}MoO_3$ powder

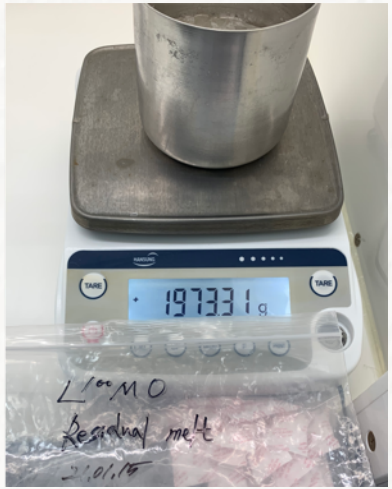
HPGe	Unpurified #3172 (CC1, 1.6 kg, 14 days)	Unpurified #3434 (CAGe, 9.6 kg, 75 days)	Unpurified #3675 (CAGe, 9.8 kg, 93 days)	Unpurified #3824 (CAGe, 12.7 kg, 183 days)	Purified #3824 (CAGe, 12.0 kg, 152 days, preliminary)
^{228}Ac	< 1.0 (90% C.L.)	0.88 ± 0.13	0.703 ± 0.097	**0.274 ± 0.044**	**< 0.030** (90% C.L.)
^{228}Th	< 1.0 (90% C.L.)	0.669 ± 0.089	0.773 ± 0.093	**0.234 ± 0.040**	**< 0.026** (90% C.L.)
^{226}Ra	5.1 ± 0.4 (stat) ± 2.2 (syst)	1.19 ± 0.42	< 0.51 (90% C.L.)	**0.273 ± 0.036**	**0.069 ± 0.014**
^{40}K	< 16.4 (90% C.L.)	36.0 ± 4.1	17.5 ± 2.0	**8.8 ± 1.0**	**1.48 ± 0.27**
^{88}Y	Not observed	0.101 ± 0.016	0.090 ± 0.013	**0.0564 ± 0.0067**	**Not observed**
^{88}Zr	Not observed	Not observed	Not observed	**0.0354 ± 0.0074**	**Not observed**

Unit: mBq/kg (kg: powder)

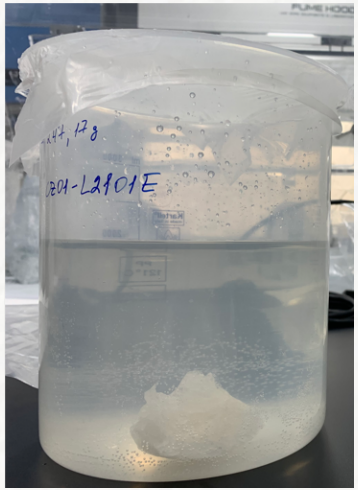
Reduced more than 4 times

ICP-MS	Unpurified #3172	Unpurified #3434	Unpurified #3675	Unpurified #3824	Purified #3824
^{232}Th	0.14	0.36	0.37	0.36	< 0.041
^{238}U	0.94	2.7	0.93	0.96	< 0.087
^{40}K	9.0	38	22	22	3.6 – 18.3

$^{100}MoO_3$ recovery from LMO melt



Residual LMO melt in the Pt-crucible



LMO melt dissolution

- 3 LMO crystal ingots are pulled from every crucible.
- The final melt is dissolved in DI-water, and insoluble impurities (Th, U, Pb, etc.) are filtered out with a membrane filter.
- Mo is separated from Li in form of molybdic acid (H_2MoO_4) via interaction with NH_4Cl.
- Separated molybdic acid is dissolved, purified with co-precipitation and recrystallized.

Recycling of the recovered $^{100}MoO_3$ powder

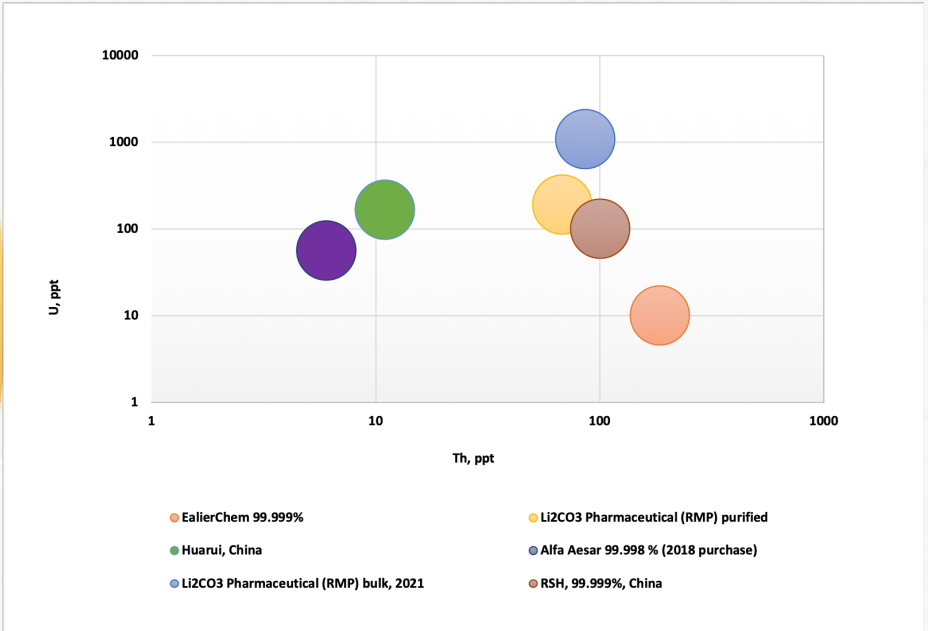
	Al	K	Ca	Ba	Sr	Pb	Th	U
	(ppb)	(ppb)	(ppb)	(ppb)	(ppb)	(ppb)	ppt	ppt
LMO melt	1450	606	579	16	0.67	<0.4	<8	28
$^{100}MoO_3$ powder, **pure recovered**	<100	131	<200	<3	<0.15	<0.4	<10	<7
$^{100}MoO_3$ powder, **initial purified**	<100	146	-	<3	<0.15	<0.4	<10	<7

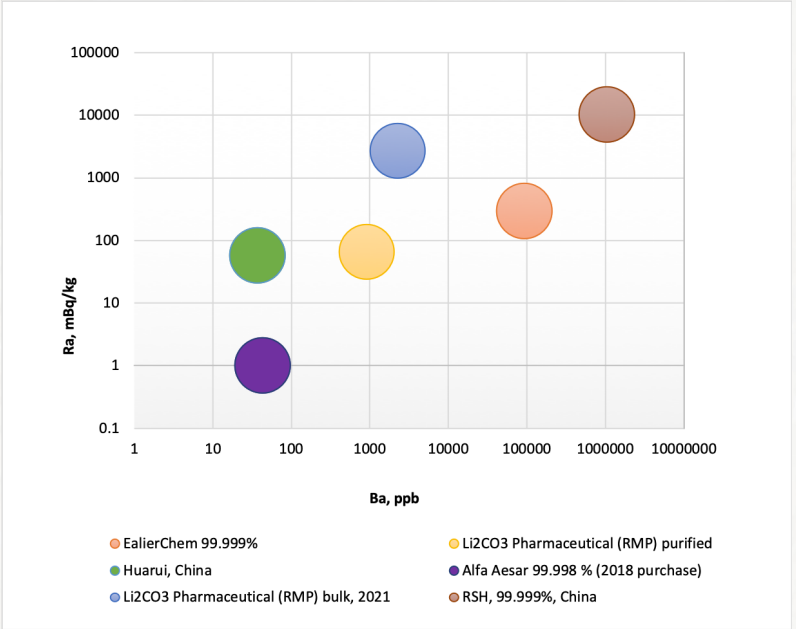
- Recovered $^{100}MoO_3$ powder is waiting for the HPGe array measurements.
- ~99% recovery efficiency for the process

02 Li_2CO_3

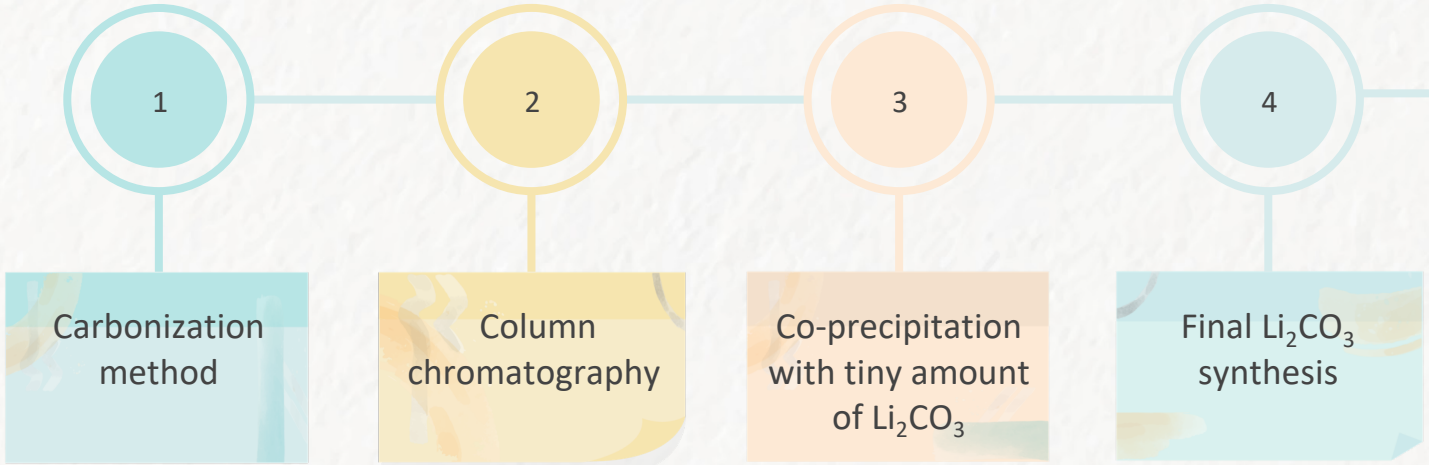
Initial material selection, purification

- Radium, but not potassium, is our enemy to defeat.
- We failed to find pure enough commercial material.
- Huarui and RMP powders were chosen as precursors for further purification at CUP.





R&D on Li_2CO_3 purification at CUP



Impurities reduction for Li_2CO_3 powder

sample	^{40}K, mBq/kg	^{228}Ac, mBq/kg	^{228}Th, mBq/kg	^{226}Ra, mBq/kg	K, ppb	Sr, ppb	Ba, ppb	Pb, ppt	Th, ppt	U, ppt
Huarui, 99.999%	HPGe@CUP				ICP-MS@CUP					
Initial	<16.59	6.33 ± 1.28	5.47 ±0.67	57.38 ± 3.15	426	6	35	1397	8	156
PURIFIED	<6.02	<1.96	<1.86	<1.1	<60	0.5	61	<80	<6	<6
DF		>3	>2	>50	>7	>12		>17	>1.3	>26

>2 **^{228}Th**

>3 **^{228}Ac**

>50 **Ra**

>7 **K**

>17 **Pb**

>26 **U**

>1.3 **^{232}Th**

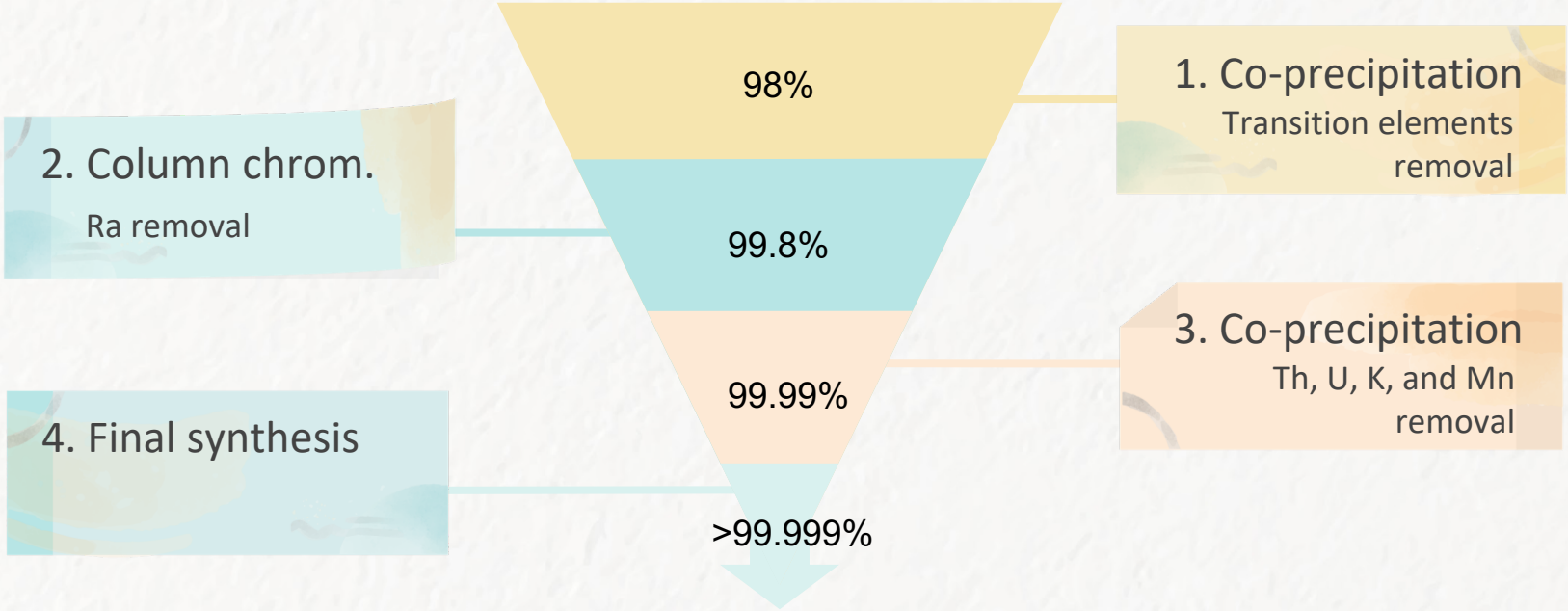
03

$CaCO_3$

Purification, recovery and recycling

	Al	K	Sr	Ba	Pb	Th	U
$CaCO_3$	(ppm)	(ppm)	(ppm)	(ppm)	(ppb)	(ppb)	(ppb)
Enriched $^{40}CaCO_3$	<2.0	<21.0	24.0	23.7	210	<0.8	<0.2
Initial 99.95%	81.5	20.3	207.2	2.3	512	33.1	878.0
Purified at CUP (2018)	11.1	1.45	175.6	1.6	5	<0.1	0.55
Purified at CUP (2019)	1.9	1.4	198.8	1.4	11	<0.1	<0.1
Purified at CUP (2020)	1.0	0.22	191	0.02	25	<0.1	<0.1
Purified at CUP (2021)		**0.21**	**204**	**0.007**	**10.2**	**0.014**	**0.036**
D.F.		*97*	*1*	*328*	*51*	*2364*	*24388*

R&D on $CaCO_3$ purification



Final $CaCO_3$ product

Summary

1. Methods for purification of $^{100}MoO_3$, Li_2CO_3, and $CaCO_3$ powders are being developed at CUP, IBS.
2. $^{100}MoO_3$ purification at CUP is going smoothly. Over 90kg of powder was treated using the developed method. The purity of produced powders is confirmed with an ICP-MS and HPGes.
3. Purification method for Li_2CO_3 powder using carbonization technique and column chromatography is under extensive study. With the current technique, over 30kg of powder was produced at CUP in the last year.
4. For the $CaCO_3$, the purification method development is ongoing. Big improvement was achieved on Ba (Ra tracer), Th, and U in the last year.
5. Methods for extraction, recovery, and purification of enriched MoO_3 and $CaCO_3$ from melt (LMO and CMO, respectively) were developed at CUP. The recovery efficiency for the methods is about 99%.

Thanks!