Institute for Basic Science

Center for Underground Physics

MoO_3 and NaI powder purification for AMoRE and COSINE to make ultra-low background crystals

Gileva Olga, HyangKyu Park, JunSeok Choe, KeonAh Shin, Pabitra Aryal, Sujita Karki

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Part 1

- **The AMoRE** (**A**dvanced **Mo** based **R**are process **E**xperiment) is searching for neutrinoless double beta decay of ^{100}Mo using radiopure molybdenum-based crystals.

AMoRE II

- $^{dep48}Ca^{100}MoO_4$ crystals: $^{dep48}CaCO_3$ & $^{100}MoO_3$ powders
- Li_2MoO_4 crystals: Li_2CO_3 & $^{100}MoO_3$ powders
- $Na_2Mo_2O_7$ crystals: Na_2CO_3 & $^{100}MoO_3$ powders

Samples	^{232}Th	^{238}U	^{226}Ra (U)	^{224}Ra (Th)	^{40}K
	ppt		mBk/kg		
^{100}Mo (99.997%)	< 46	73	8.3	< 1	9
	< 61	149	3.8	< 0.8	36
^{dep48}Ca	< 1000	< 1000	51	1	-
Li_2CO_3 (99.998%)	9.6	414	0.95	0.41	9.0
Na_2CO_3 (99.997%)	< 52	< 52	4.15	0.52	31.5

Requirements for ^{238}U & ^{232}Th:

~ μBq/kg in crystals ~ 1 000 reduction

AMoRE purification experiments

- Purification of raw MoO_3 powder:
  - sublimation (99% efficiency, no need another chemicals)
  - fractional recrystallization (minimal irretrievable losses, no need another chemicals, achieved efficiency after one step is 30-60 %, purity level depends on efficiency)
  - 3% co-precipitation with $CaCl_2$ (around 3% losses of Mo, necessary to find very pure carrier, not less 99.999% grade)

- Preparing of final product:
  - precipitation of PAM (polyammonium molybdate) by HCl (90-95 % efficiency, PAM powder contain NH_4Cl and HCl)
  - precipitation of PAM by complete evaporation (irretrievable losses around 1 %, procedure takes time)

PAM – PolyAmmonium Molybdate $(NH_4)_2\ Mo_4O_{13}{\times}2H_2O$

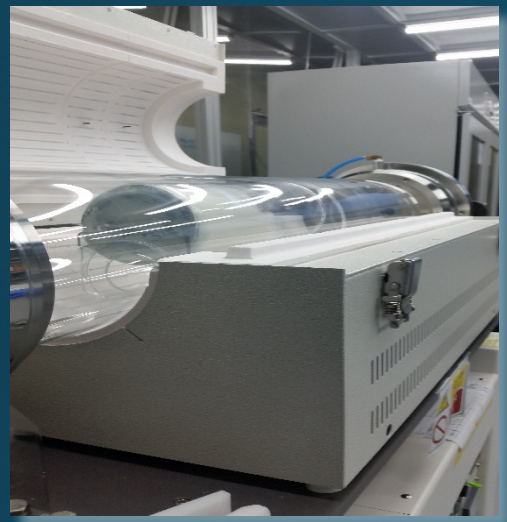
Purification of raw MoO_3 powder

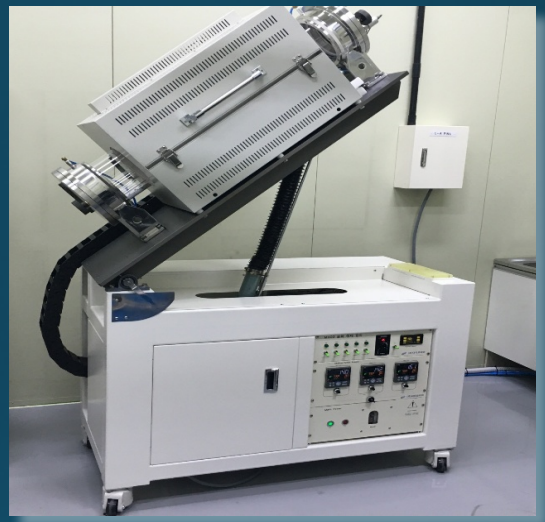
Purification technique	Sr, ppt	Ba, ppt	Pb, ppt	Th, ppt	U, ppt
	Decontamination factors (DF)				
Initial raw powder	6624	1366580	16014	270	4108
Fractional recrystallization	61.3	0.57	23.4	1.82	27.2
Initial MoO_3 powder	23714	2829035	107027	91	7014
Co-precipitation with $CaCl_2$	3 (64%)	217 (99%)	61 (98.5%)	≥1	31 (97%)
Evaporation of solvent	0.44	0.44	≃ 1	≃ 1	≃ 1
Precipitation of PAM with HCl	≃ 100	1.9	62.9	>2.4	9

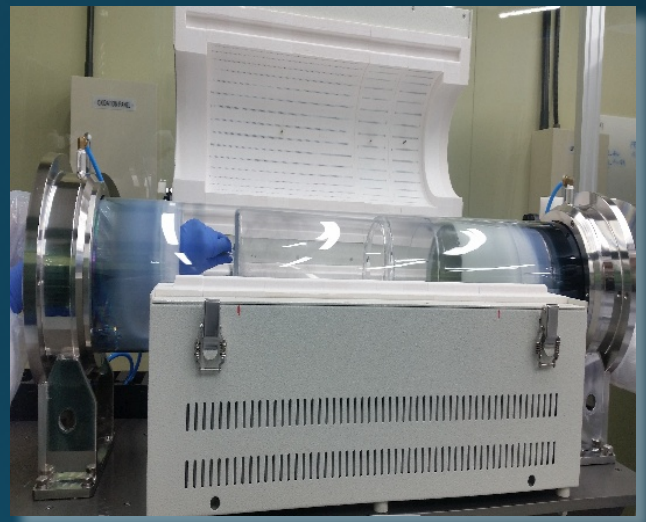
Combined methods by wet chemistry technique

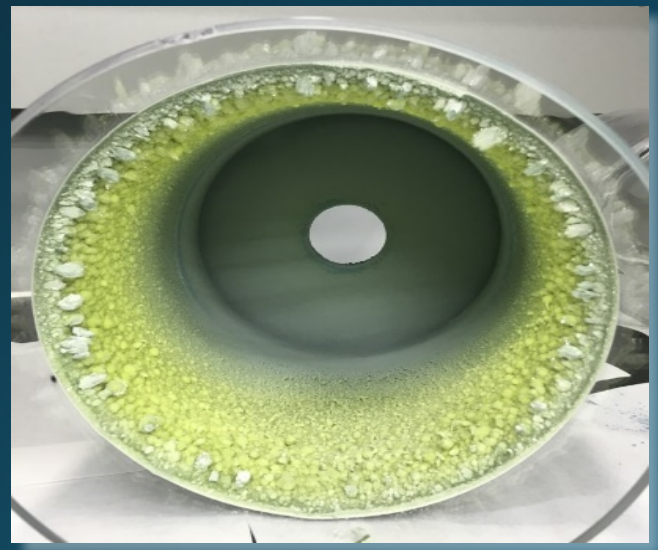
Description	Sr, ppt	Ba, ppt	Pb, ppt	Th, ppt	U, ppt
1. Initial MoO_3 → co-precipitation → evaporation of solvent → PAM powder					
Initial MoO_3 powder	23714	2829035	107027	91	7014
DF after co-precipitation	3.1	228	78	1.4	14
DF after evaporation	0.5	57.7	59.7	0.63	6.2
2. Initial MoO_3 → co-precipitation → precipitation with HCl → PAM powder					
Initial MoO_3 powder	23714	2829035	107027	91	7014
DF after co-precipitation	3	217	61	≥1	31
DF after precipitation with HCl	310.4	221	476.5	≥3.5	≥200

Vacuum sublimation procedure









- Loading of MoO_3 powder in quartz tube
- Loading quartz tube in sublimation machine
- Operating the machine at 720 °C and vacuum condition (<10 mtorr)

Sublimation of initial MoO_3 powder

Description	Sr, ppt	Ba, ppt	Pb, ppt	Th, ppt	U, ppt
Initial powder	23714	2829035	107027	91	7014
Single sublimation	930	123076	68853	<50	612
Decontamination Factor	25	23	2	>2	11
Double sublimation	89	17084	60296	<35	66
Decontamination Factor	266	166	2	>2	106

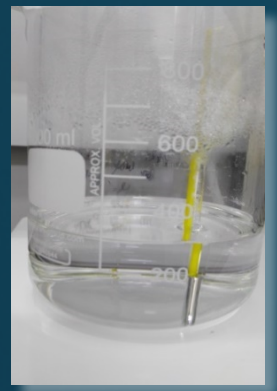
Efficiency after sublimation is more 99% !

Correlation of contamination level of MoO_3 powder obtained by ICP-MS and HPGe

Samples	Ba	^{232}Th	^{238}U	^{226}Ra (U)	^{228}Ac (Th)	^{40}K
		ppt			mBq/kg	
$^{100}MoO_3$ (99.997%)	16600	< 46	73	8.3	< 1	9
	11400	< 61	149	3.8	< 0.8	36
Ini. MoO_3	2829035	91	7014	1627	851	725.48
Single Sub.	123076	< 50	611.8	124.7	49.9	73.84
Double sub.	17084	< 35	66.09	20.56	6.73	74.80

Total Reduction Factor for ^{226}Ra $\sim$ 80, for ^{228}Ac $\sim$ 125, for ^{40}K $\sim$ 10

Upgrading of evaporation technique

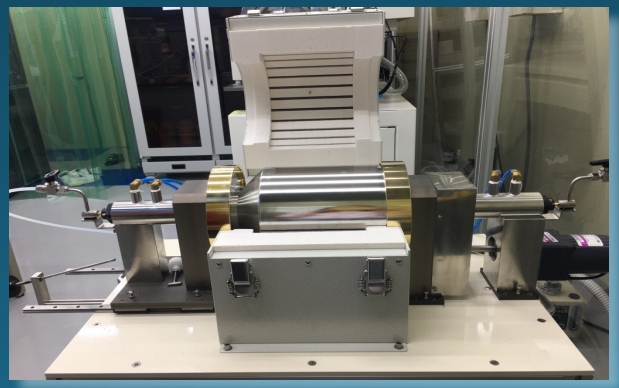


Evaporation on the heating plate at 70 °C

Rotary evaporation at 70 °C in vacuum condition

Annealing technique

Purified PAM powder



Annealing at 675°C

MoO_3

Part 2

- **The COSINE** collaboration (**C**ollaboration **O**f **S**odium **I**odi**N**e **E**xperiments) is searching for dark matter with ultra-low background NaI crystals either to confirm or to refute the DAMA signal.

COSINE-100

- K: NaI powder should be purified up to level below **10** ppb

Goal : confirm or exclude the DAMA/LIBRA's modulation results

Strategy : achieve ultra-low background

How to purify NaI powder?

- Fractional recrystallization from water solution (water as solvent is very good, low yield after single procedure, takes time, labour-consuming)

- Solvent extraction (need to find very pure extractants, not effective for K)

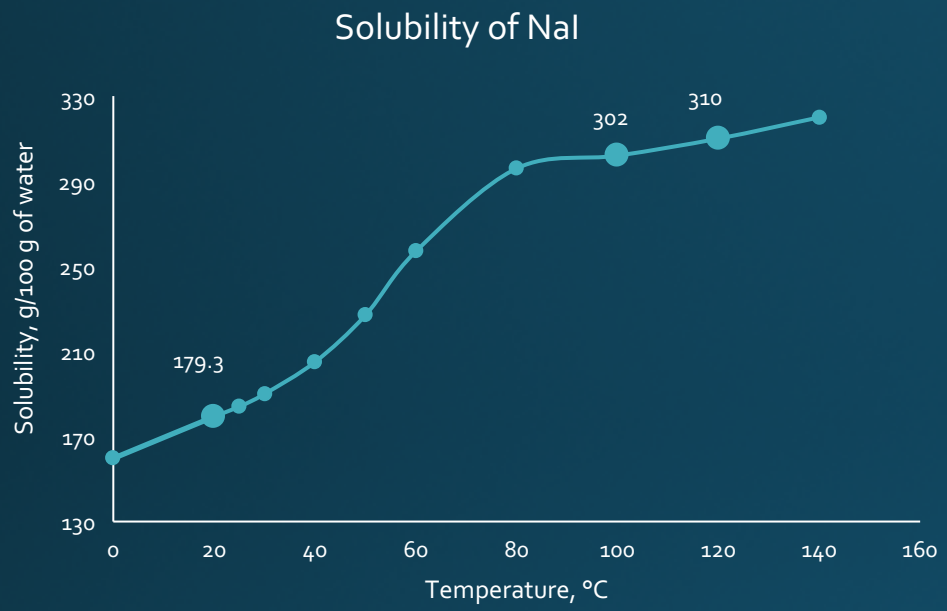
- Desalting from system acetone-water or ethanol-water (low yield after single procedure, takes time, labour-consuming, need to find very pure organic solvents)

Fractional Recrystallization of Crystal and Astro grade NaI powder

	Ba^{138}, ppb	K^{39}, ppb	Pb^{208}, ppb	Sr^{88}, ppb	Th^{232}, ppb	U^{238}, ppb
Initial CG	7.14	45.07	3.3	0.9	DL	DL
Purified CG	0.618	6.04	0.81	<0.3	DL	DL
DF	11.5	7.5	4	>3	-	-
Initial AG	0.6	4.511	0.93	<0.3	DL	DL
Purified AG	<0.3	<1.0	<0.4	<0.3	DL	DL
DF	>2	>4.5	>2	-	-	-

CG – crystal grade powder; AG – astro grade powder.
ICP-MS data from SEASTAR, Canada

Fractional recrystallization procedure for the NaI powder



Mother solution could be used as solvent for previous recrystallization step

Preparing of saturated solution at 20 °C

↓

Adding reduction agent in order to prevent I_2 and IO_3^-

↓

Filtration using membrane filter

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Evaporation of water in presence of HI

↓

Cooling down of solution and filtration

↓

Washing of obtained crystals by Ethanol

↓

Successful drying of crystals at 65 and 130 °C

Fractional Recrystallization of NaI powder

Sample	Ba^{138}, ppb	K^{39}, ppb	Pb^{208}, ppb	Sr^{88}, ppb	Th^{232}, ppb	U^{238}, ppb	efficiency
Initial NaI powder (technician grade)	2592	180000	5.674	65.676	<0.100	<0.100	-
1st recrystallized NaI crystal	25.5	6280	0.364	0.650	<0.100	<0.100	52.8 %
D.F	102	29	16	101	-	-	
2nd recrystallized NaI crystal	5.3	1305	0.151	0.152	<0.100	<0.100	27.5 %
D.F	490	138	38	432	-	-	
NaI crystals from 2nd MS	226.5	2745	0.400	1,263	<0.100	<0.100	16.7 %
D.F	11	66	14	52	-	-	

MS – mother solution
ICP-MS data from SEASTAR, Canada

summary

Using the double sublimation of MoO_3 powder we reached Total Reduction Factor for $^{226}Ra \sim 80$, for $^{228}Ac \sim 125$, for $^{40}K \sim 10$

Combined methods of Mo-purification looks very promising

We could achieve final goal using the Fractional Recrystallization of NaI powder, which has initial K contamination around 1 ppm

Our experiments are ongoing


ngiyabonga
danke
謝謝
teşekkür ederim
спасибо
dank je
tapadh leat
thank you
gracias
bedankt
hvala
mauruuru
mochchakkeram
dziękuję
go raibh maith agat
obrigado
sagolun
sukriya
kop khun krap
arigatō
takk
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grazie
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ευχαριστώ