Production of low background scintillating crystals for underground experiments in Korea

Moo-Hyun Lee

Center for underground Physics,
Institute for Basic Science, Korea

2019. 05. 21

LRT 2019 @ Palacio de Congresos, Jaca, Spain

Yangyang underground laboratory (Y2L) & CUP

(Upper Dam)

YangYang Pumped
Storage Power Plant

Center for Underground Physics
IBS (Institute for Basic Science)

1000m

700m

(Power Plant)

양양양수발전소

(Lower Dam)

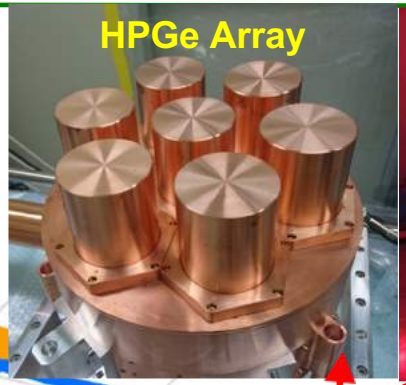
COSINE-100 (WIMP Dark Matter)

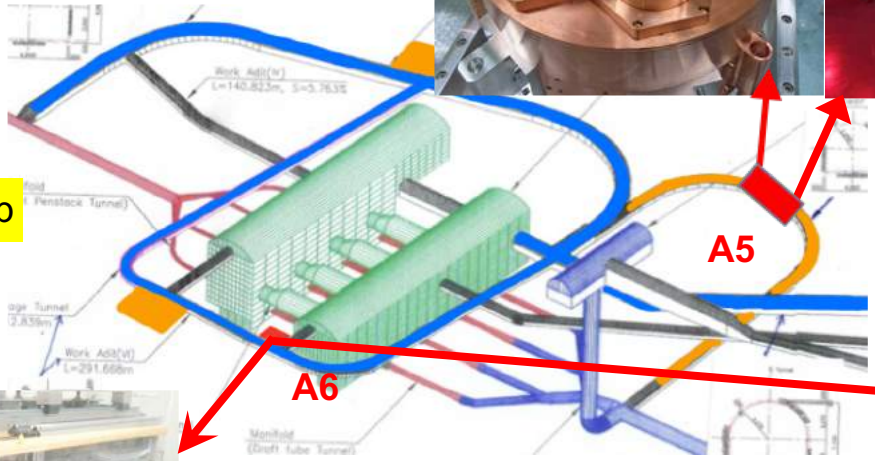
AMoRE ($0\nu\beta\beta$ Decay)

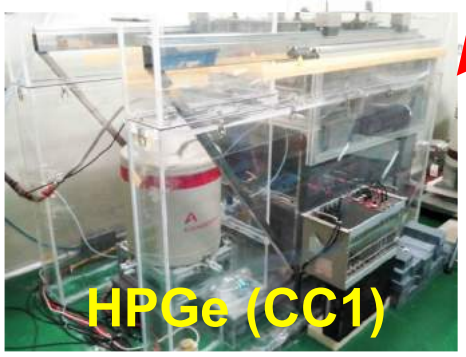
Minimum depth : 700 m / Access to the lab by car (~2km)

Low Background Measurements at Y2L

- ☐ 2 HPGe detectors (2 Coax)
- ☐ 1 Array with 14 HPGe detectors
- ☐ Alpha ionization counter

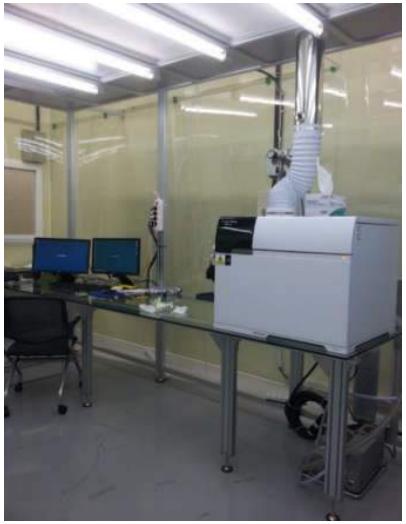






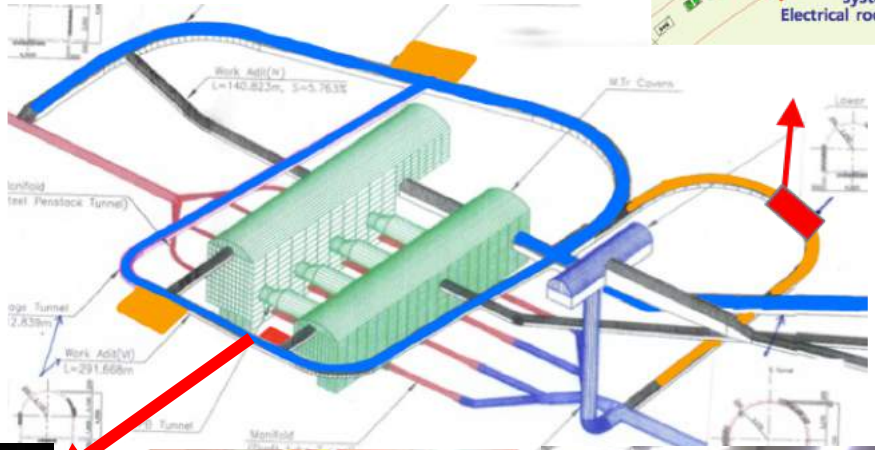
ICP-MS Lab at IBS HQ, Daejeon

- Agilent 7900, the highest sensitivity single MS system in 2015 when purchased.
- Under operation since Oct. 2015. (Moved to IBS HQ in spring 2018)
- In a cleanroom nominally designed as class 1000, >150 air changes/hour.
- A Millipore DI system, in-house acid distillation with a 3 linear meters of chemical hood space.
- Dissolve sample in liquid form, uptake in argon (Ar) gas stream, ionize gas, extract into mass spectrometer, measure trace contaminants.
- Confirmation of purification methods by measuring isotopic or chemical tracers.
- Confidence in systematics at ultra-trace levels is not easily achievable through outsourced measurements.



Experiments at the Y2L

- New rooms in A5 tunnel for COSINE (38 m^2) and AMoRE (43 m^2) constructed in late 2014.
- Currently the two experiments are running together with an array of 14 HPGe.



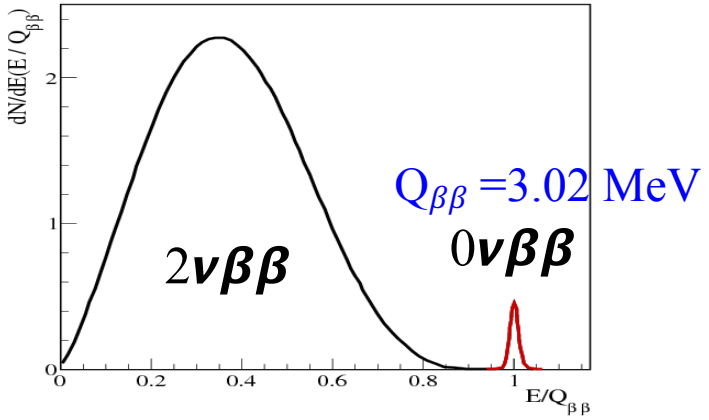
AMoRE Experiment: 0νββ decay search

CENTER FOR UNDERGROUND PHYSICS ibs

● Sizable background case:

$$T_{1/2}^{0\nu}(\exp) = (\ln 2) N_A \frac{a}{A} \varepsilon \sqrt{\frac{Mt}{b\Delta E}}$$

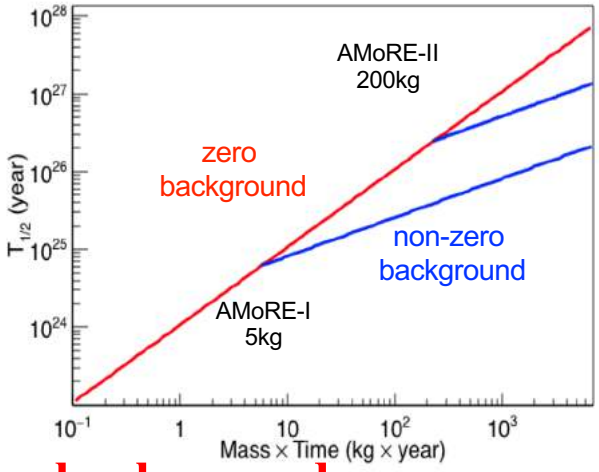
- Isotopic Abundance
- Detection Efficiency
- Detector Mass
- Measurement time
- Energy Resolution
- Sensitivity to half-life of 0νββ
- Atomic mass
- Background rate



● "Zero" background case:

When b is ~ O(1),

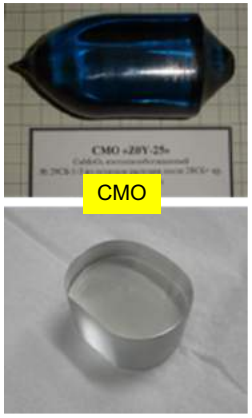
$$T_{1/2}^{0\nu}(\exp) = (\ln 2) N_A \frac{a}{A} \varepsilon M t$$

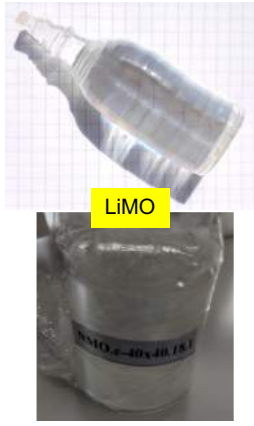


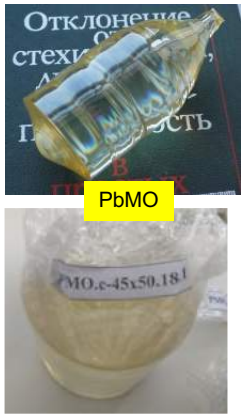
AMoRE is aiming for zero background.

AMoRE Parameters

- Crystals: $^{40}Ca^{100}MoO_4$(CMO) or XMO
  (X: Li, Na, or Pb)
  - ^{100}Mo enriched: > 95%
  - ^{48}Ca depleted: < 0.001% (N.A. of ^{48}Ca:0.187%)
- Low temperature detector: 10 – 30 mK
- Energy resolution: ~5 keV @ 3MeV
- Excellent PSD







The AMoRE configuration and plan

	Pilot	Phase I	Phase II
Mass (Crystal)	**1.9 kg CMO**	**6 kg (CMO + LMO)**	**200 kg XMO (X: Li, Na, Pb)**
BKG [keV·kg·year]$^{-1}$	**<10^{-2}**	**<10^{-3}**	**<10^{-4}**
$T_{1/2}$ Sensitivity [years]	**~10^{24}**	**~10^{25}**	**~8 ×10^{26}**
<$m_{\beta\beta}$> Sensitivity [meV]	**400 - 700**	**100 - 300**	**13 - 25**
Location	**Y2L (700 m depth)**		**Yemi Lab (1100m depth)**
Schedule	**2015 - 8**	**2019 - 2021**	**2021 -**

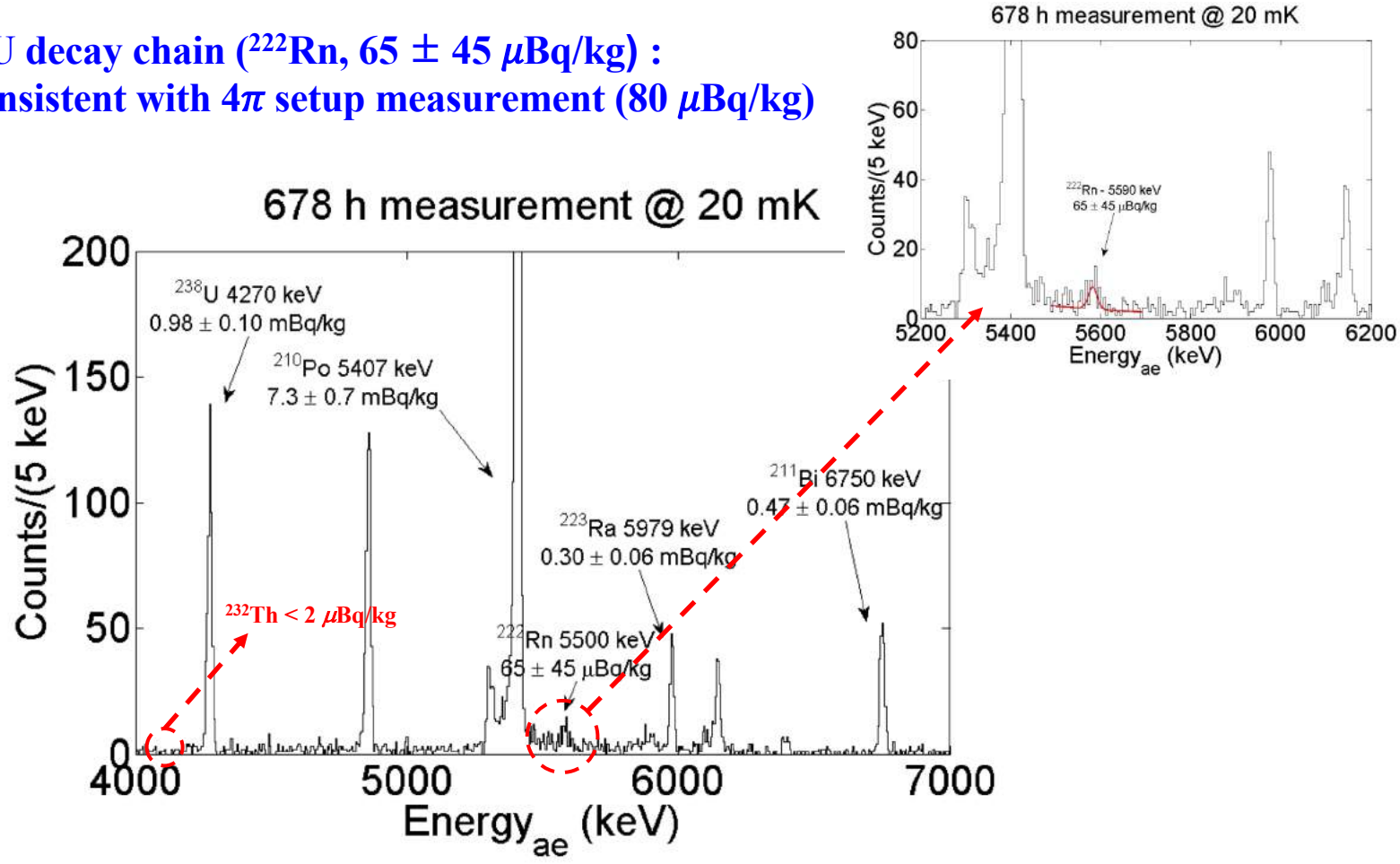
Identify critical radioactivity

- Go through all known nuclei decaying β with $Q_{\beta\beta} > 3.02$MeV in NNDC database.
- ^{110m}Ag (3.0105 MeV) doesn't contribute for Mo experiment.
- Cosmogenic excitation is negligible after 1 year at underground.
- Only Th and U natural radio-activities are critical for Q> 3.02MeV. → Great advantage to run high Q-value nuclei !

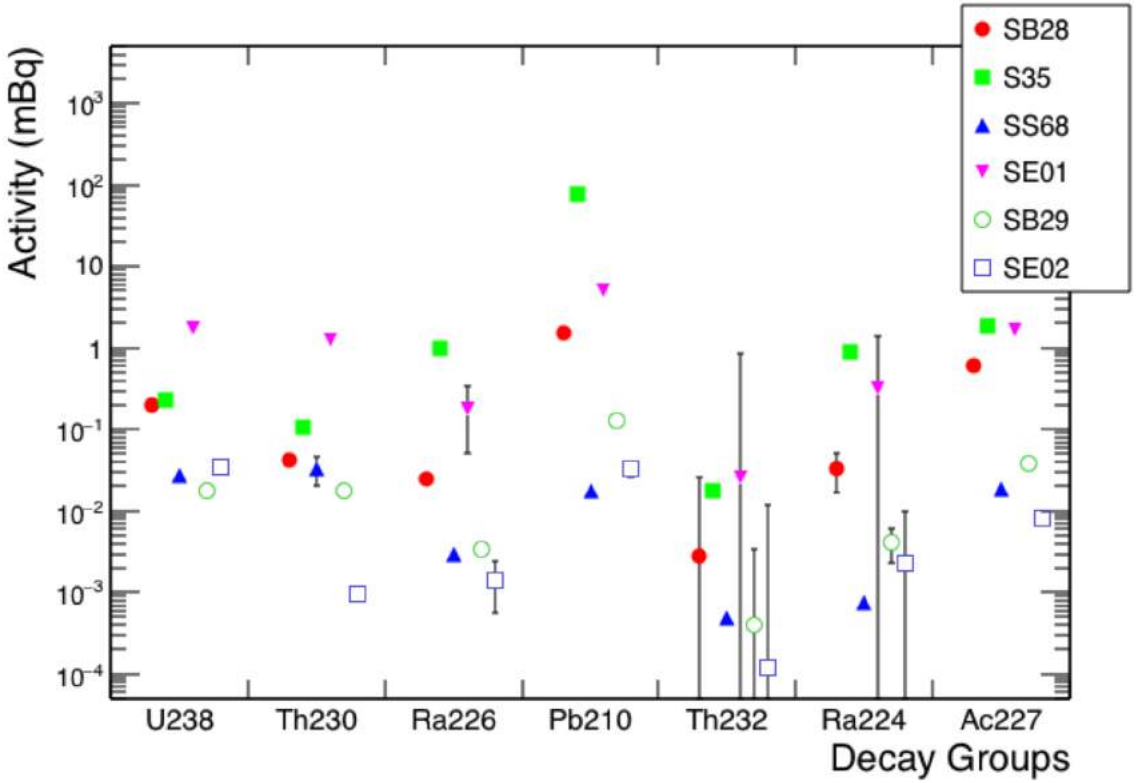
Element	Decay	$T_{1/2}$	Q (MeV)	Mother	Chain	Comment
^{26}Al	EC	7.4x10^{5}y	4.004	N/A		Long lifetime
^{56}Co	EC	0.21y	4.567	N/A		Short lifetime
^{88}Y	EC	0.29y	3.623	^{88}Zr (0.23 y)		Short lifetime
^{106}Rh	B-	30s	4.004	^{106}Ru(1.02y)		
^{126}Sb	B-	12.5d	3.670	^{126}Sn(2.3x10^{5}y)		Long lifetime
^{146}Eu	EC	4.61d	3.878	^{146}Gd (0.13 y)		Short lifetime
^{208}Tl	B-	3.05m	4.999	^{228}Th (1.91 y)	^{232}Th	Main
^{209}Tl	B-	2.16m	3.970	^{233}U(159200y)	^{233}U	2.1% branching
^{210}Tl	B-	1.3m	5.482	^{226}Ra(1600y)	^{238}U	0.02% branching
^{214}Bi	B-	19.9m	3.269	^{226}Ra(1600y)	^{238}U	Main

Internal alpha background of SB28 (AMoRE-Pilot)

- ^{238}U decay chain (^{222}Rn, 65 ± 45 μBq/kg) :
  Consistent with 4π setup measurement (80 μBq/kg)



AMoRE-Pilot: Internal Activities from α rates



- Most of the ^{210}Pb are bulk contribution.
- Internal backgrounds between crystals differ more than an order, even two orders.

AMoRE-I CMO crystals (FOMOS)

Total: 3.387 kg

- 3 years for procurement.
- LY, resolution, transmittance, RT background measurements done.
- #3-9 to be installed at AMoRE-I together with #1-2.

AMoRE-I CMO: RT measurement

[μB/kg]	^{227}Ac (^{215}Po) (^{235}U family)	^{226}Ra (^{214}Po) (^{238}U family)	^{228}Th (^{216}Po) (^{232}Th family)	Alpha	Relative Light Yield
Qualification*	<500	<100	<50	<1000	
SE1	60 ± 8	40 ± 6	50 ± 6		0.43
SE2	90 ± 10	20 ± 3	< 100		0.58
SE3	30 ± 6	6 ± 3	30 ± 6	28000	0.75
SE4	30 ± 6	10 ± 3	10 ± 3	3200	0.60
SE5	40 ± 6	10 ± 3	10 ± 3		0.70
SE6	35 ± 6	100 ± 10	70 ± 10		0.62
SE7	80 ± 10	30 ± 5	65 ± 10		0.60
SE8	40 ± 6	20 ± 5	40 ± 6		0.55
SE9	0 ± 6	<11	50 ± 6		0.66

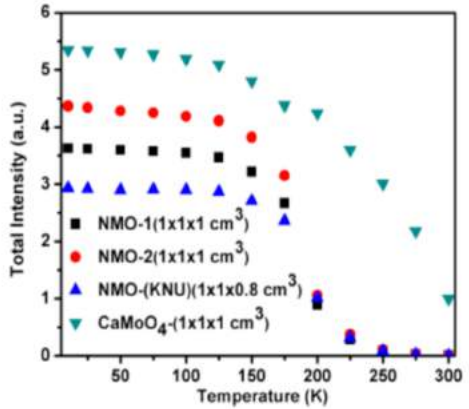
***Expect to have 10^{-3} counts/keV/kg/y at AMoRE-I**

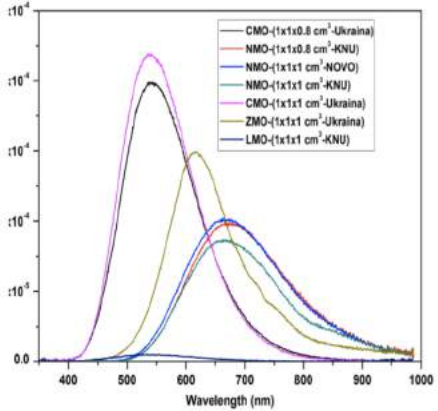
J.Y. Lee et al., IEEE TNS vol. 65 No. 8 (2018) 2041

Decision on crystals for AMoRE-II

- CMO ($CaMoO_4$) is a very good crystal with the largest light output, but CMO has a disadvantage that we need ^{48}Ca depleted isotopes, expensive.
- CUPID-Mo group decided to use LMO (Li_2MoO_4), and we are working on LMO, PMO ($PbMoO_4$), & NMO ($Na_2Mo_2O_7$), crystals.

Crystal	Emission (nm)	LightYield (@10K)		Decay time (μs)	density	Mo Fraction
		280nm	X-ray			
CMO(Ukra)	540	100	100	240	4.34	0.49
ZMO(NIIC)	614	63	35		4.37	0.436
LMO(KTI)	535	1	5	23	3.03	0.562
PMO(NIIC)	592	11	105	20	6.95	0.269
NMO(NIIC)	663	75	9	750	3.62	0.558





- CMO ($CaMoO_4$)
- LMO (Li_2MoO_4)
- NMO ($Na_2Mo_2O_7$)
- PMO ($PbMoO_4$)

Pandey et al., IEEE Trans. Nucl. Sci. (2018)
Pandey et al., Journal of Crystal Growth 480 (2017) 62-66
J.Y. Lee et al., IEEE Trans. Nucl. Sci. (2018)

Ultra-low background crystals for AMoRE-II

Ultra-low background powder R&D is difficult and need **quick feedback**
(Purification and measurement of 10 μBq/kg ^{238}U, ^{232}Th & total radioactivity of alpha < 1 mBq)

CUP chemical & clean room

CUP & underground lab

CUP lab.

Y2L underground lab
Baksan lab.

Company
(CUP lab.)

Powder purification

ICP-MS, HPGe, ...

No

Yes

Re-crystalization

Crystal growing

Scintillation
(Phonon)

No

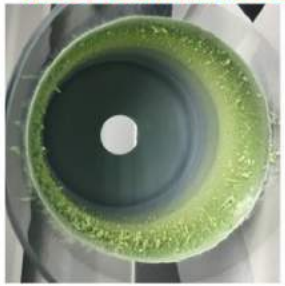
Yes

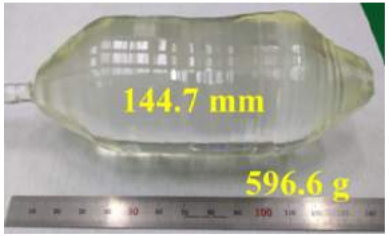
Crystal growing optimization
(Size and quality)

Low background Crystal growing facility at CUP

- Main goal
  - $CaMoO_4$, Li_2MoO_4, $Na_2Mo_2O_7$ crystals growing R&D for AMoRE-II
  - Other DBD or DM crystal R&D
- Deep purification of $CaCO_3$ and MoO_3 powders (< 50 μBq/kg for U,Th chain)
  (Details in Olga Gileva's talk)
- Crystal growing equipment:
  3 Czochralski, 2 Kyropoulous, 1 Bridgman crystal growers.

Purified powder after sublimation

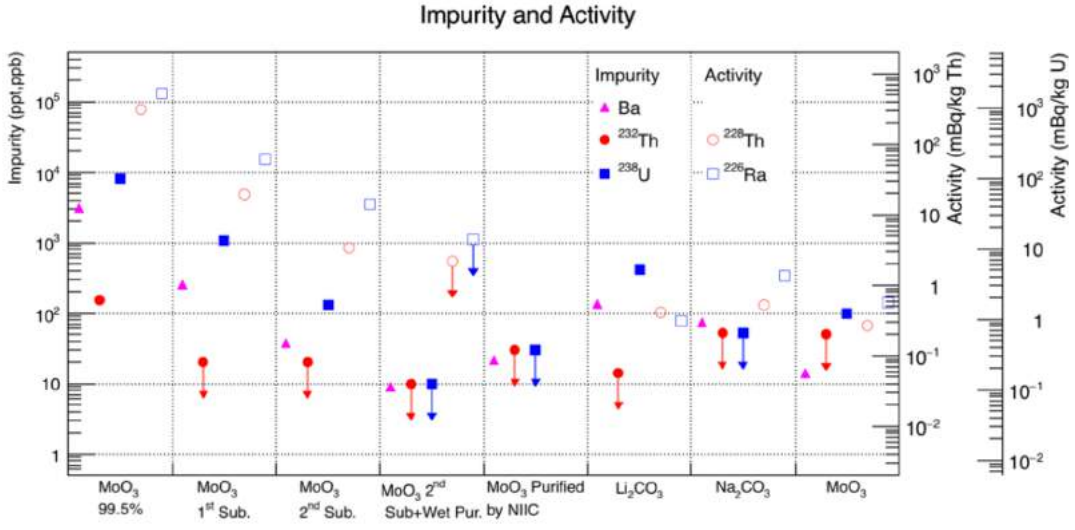


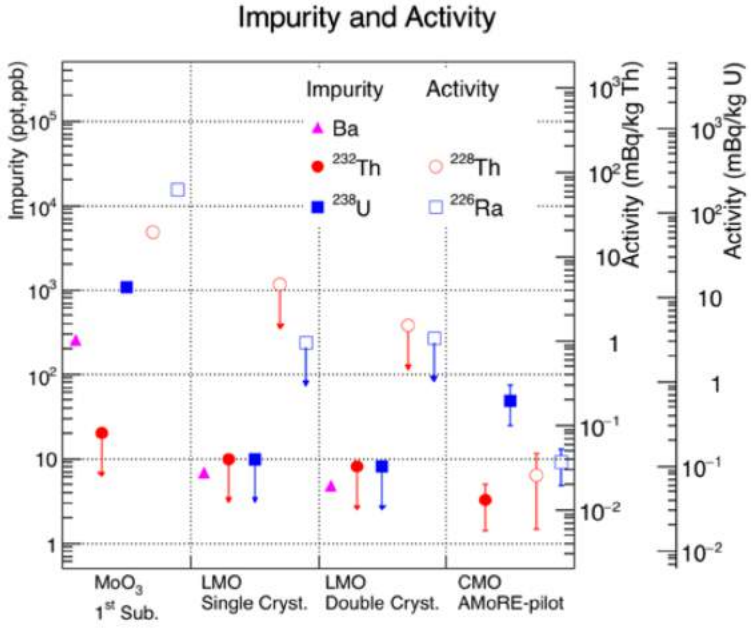


CMO & LMO crystals by CUP

AMoRE-II: Purification of XMO crystals

- Ba is a good indicator for Ra since they are in the same family.
- We have a good progress toward AMoRE-II crystals.





Details in Olga Gileva's talk

Molybdate crystals grown at CUP

- Successful in growing molybdate crystals. Growing time ~ 1 week.
- The purity of the grown crystals are measured by ICP-MS → Promising results
- Enriched LMO crystals are grown at NIIC and CUP for LT measurements.

Unpurified Mo and Ca powders

Purified Mo and Li powders

Purified Mo and Na powder

255.6 g, 100 mm (L)

cut and polished

Purities of CUP grown LMO crystals

CENTER FOR UNDERGROUND PHYSICS ibs

Element		Al	K	Ba	Sr	Pb	Th	U
No.	sample	(ppb)	(ppb)	(ppb)	(ppt)	(ppt)	(ppt)	(ppt)
Single crystallized natural LMO (w/o purification)								
CMD 113	L1701-1	48.1	347.3	5.445	<15	<300	<15	<16
CMD 113	L1701-2	21.7	449.2	5.401	75	<300	<15	<16
Single crystallized natural LMO (MoO_3 sublimed)								
CMD163.1	CZ02-L1706-T	<11	38	7.579	<50	<100	<8	<8
CMD163.2	CZ02-L1706-B	<11	83	9.617	<50	<100	<8	<8
Double crystallized natural LMO (MoO_3 sublimed)								
CMD191.1	CZ02-L1801-T	<11	<30	4.744	<50	<100	<8	<8
CMD191.2	CZ02-L1801-B	<11	<30	5.814	<50	<100	<8	<8
Enriched LMO (w/o purification)								
CMD00236.2	CZ02-L1803E-T	1437	<40	6.82	<31	<225	<6	<6
CMD00236.3	CZ02-L1803E-B	1484	<40	7.07	<31	<225	<6	<6
CMD00236.1	CZ02-L1803E-RM	3824	249	28.58	4110	12290	71	472

Results for a natural CMO crystal growth at CUP

CENTER FOR UNDERGROUND PHYSICS ibs

No.	Sample	K, ppb	Al, ppb	Cr, ppb	Fe, ppb	Sr, ppb	Ba, ppb	Pb, ppb	Th, ppb	U, ppb
Initial MoO_3 99.95%						6.6	1366	16.0	0.3	4.1
Initial $CaCO_3$ 99.95%						162936	3081	449	50	1429
CMD92 (CZ01-1604)	Top	439	<60	24	1475	59413	116	6.6	1.2	9.3
	Middle	513	<60	18	1756	62315	134	6.5	1.3	9.9
	Bottom	449	<60	13	2051	69216	156	5.4	2.1	10.4

Mo-100 powder (ECP) for AMoRE-II

Contract Date (weight)	Lot#	Delivery@Y2L
2015 (10 kg)	#1(3172)	3/9/16
	#2(3328)	
2016 (10 kg)	3434	12/28/16
2016 (10 kg)	3497	10/12/17
2016 (10 kg)	3535	
2017 (10 kg)	3589	2/7/18
2017 (10 kg)	3649	5/29/18
2017 (10 kg)	3675	8/14/18
2017 (10 kg)	3741	2/13/19

80 kg delivered out of 110 kg

Last delivery ~Sep. 2019

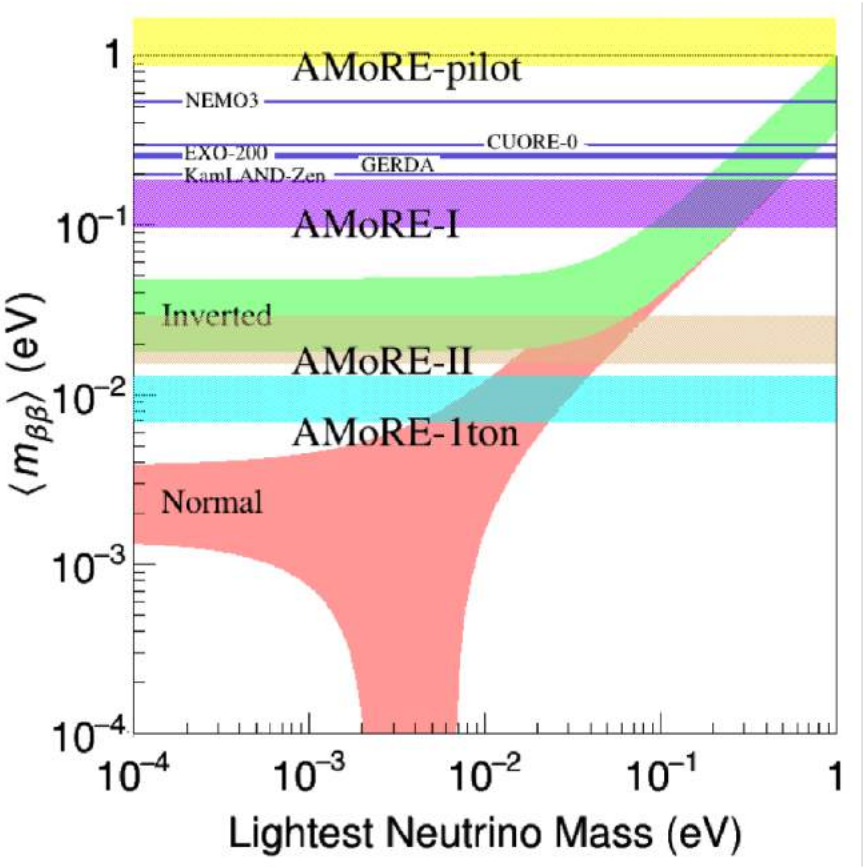
HPGe Array meas. (9/13 – 11/28/2017)

- ^{226}Ra chain (^{238}U):1.6 ± 0.3 mBq/kg
- ^{228}Th chain (^{232}Th): 244 ± 50 μBq/kg (first measurement)
- ^{88}Y: 33 ± 8 μBq/kg (cosmogenic)

AMoRE sensitivity for neutrino mass

- Seven commissioning runs in AMoRE-pilot have been completed in December 2018.
- AMoRE-I is currently being prepared to start from fall this year.
- AMoRE-II preparation is ongoing in parallel together with the Yemilab construction.

  - Nuclear Matrix Element: QRPA (Faessler et al., 2012)
  - AMoRE-I: 5 kg and 5 years
  - AMoRE-II: 200 kg and 5 years
  - It was assumed as "zero-background".



COSINE-100 instrument for WIMP DM search

JINST 13 T02007 (2018)

4π Muon Counter
37 plastic scintillator panels
2-inch PMT(H7195)s for muon counter

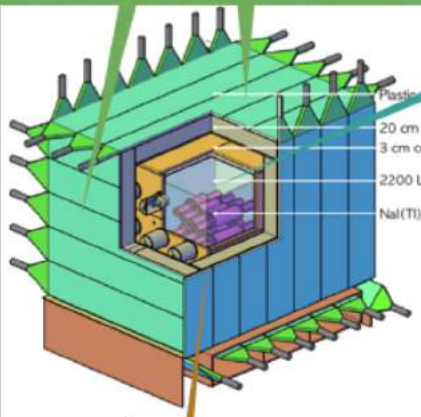
Nucl. Instrum. Meth. A 851 103 (2017)

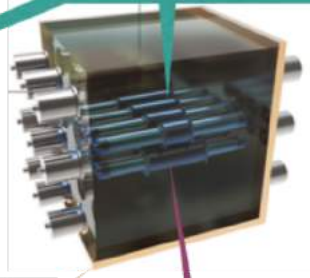
Liquid Scintillator
2200-L LAB-based LS for veto
5-inch PMT(R877)s for LS detector

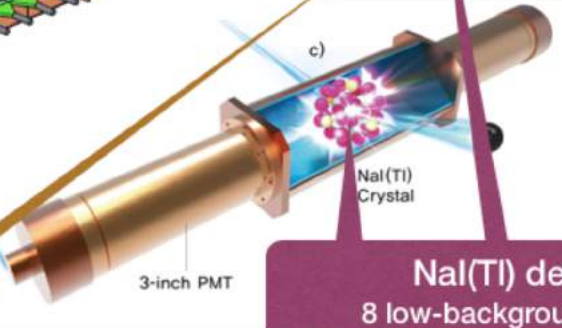
JINST 13 T06005 (2018)

Neutron Monitoring
Fast neutron detector
(Liquid scintillator)

Thermal neutron detector
(^{3}He gas detector)







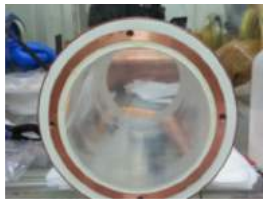
Shields
3-cm thick copper box
20-cm thick lead shielding

NaI(Tl) detector
8 low-background crystals
Each crystal is encapsulated in copper
Two 3-inch PMTs for each crystal
(R12669SEL)

Eur. Phys. J. C. 78 107 (2018)

COSINE-100 NaI(Tl) crystal detector

- 8 ultra low-background NaI(Tl) crystals with a mass of 106 kg in total
- U/Th/K levels are less than DAMA, but total alphas (^{210}Pb) are higher than DAMA.
- Higher light yield (15 p.e/keV) than DAMA
- Can make the threshold lower easily
- Total background level is 2-3 times that of DAMA.

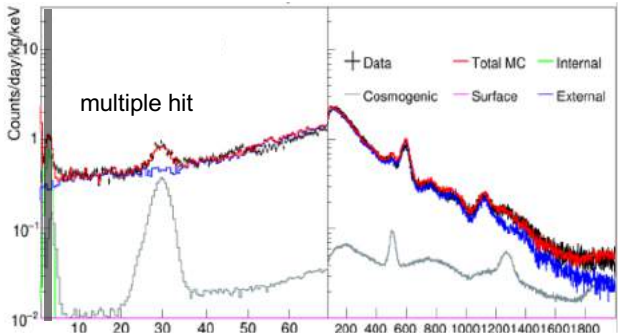


Crystal	Mass (kg)	Powder	Alpha rate (mBq/kg)	^{40}K (ppb)	^{238}U (ppt)	^{232}Th (ppt)	Light yield (p.e./keV)
Crystal 1	8.3	AS-B	3.20 ± 0.08	43.4 ± 13.7	< 0.02	1.31 ± 0.35	14.88 ± 1.49
Crystal 2	9.2	AS-C	2.06 ± 0.06	82.7 ± 12.7	< 0.12	< 0.63	14.61 ± 1.45
Crystal 3	9.2	AS-WS Ⅱ	0.76 ± 0.02	41.1 ± 6.8	< 0.04	0.44 ± 0.19	15.50 ± 1.64
Crystal 4	18.0	AS-WS Ⅱ	0.74 ± 0.02	39.5 ± 8.3		< 0.3	14.86 ± 1.50
Crystal 5	18.0	AS-C	2.06 ± 0.05	86.8 ± 10.8		2.35 ± 0.31	7.33 ± 0.70
Crystal 6	12.5	AS-WSⅢ	1.52 ± 0.04	12.2 ± 4.5	< 0.018	0.56 ± 0.19	14.56 ± 1.45
Crystal 7	12.5	AS-WSⅢ	1.54 ± 0.04	18.8 ± 5.3		< 0.6	13.97 ± 1.41
Crystal 8	18.3	AS-C	2.05 ± 0.05	56.15 ± 8.1		< 1.4	3.50 ± 0.33
DAMA			< 0.5	< 20	0.7 - 10	0.5 – 7.5	5.5 – 7.5

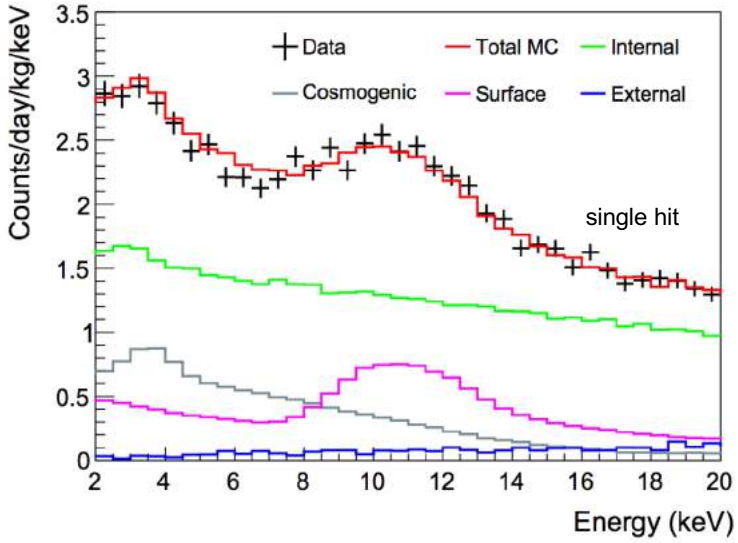
Eur. Phys. J. C. 78 107 (2018)

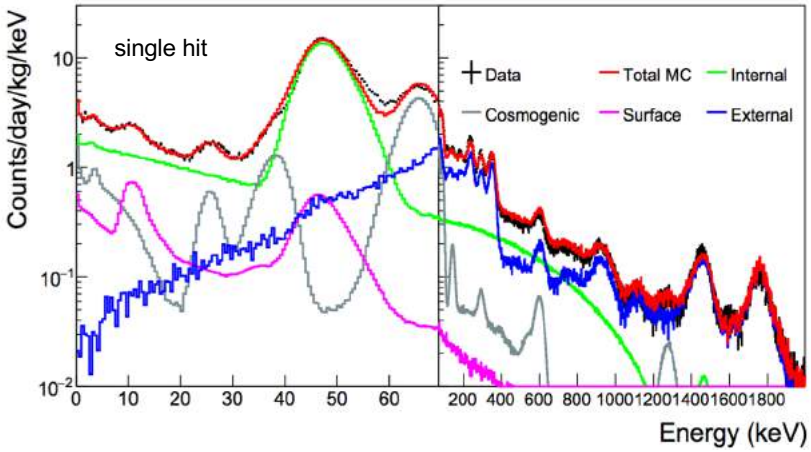
COSINE-100 Background Modeling

- Detector simulation based on GEANT4 well reproduces measured background spectra (4 channels for each crystal, single: 6 - 2000 keV, multiple: 2 - 2000 keV).
- Main background in low energy region
  - Contamination of crystals from ^{40}K/U/Th
  - Cosmogenic activation (mostly ^{3}H)
  - ^{210}Pb on crystal surface



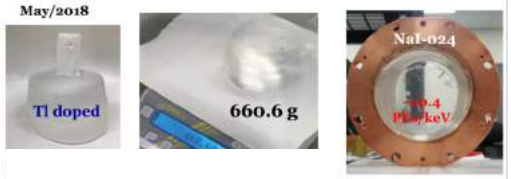
Eur. Phys. J. C. 78 490 (2018)





Next Phase of COSINE(-200)

- Extremely pure crystal development
  - Background level less than that of DAMA/LIBRA (1 dru)
  - In-house development for the entire processes
    - Purification of NaI powder
    - Full size crystal grower
- R&D of NaI(Tl) crystal at low temperature
- Current COSINE-100 shield designed to accommodate sixteen of 12.5 kg crystals (200 kg).



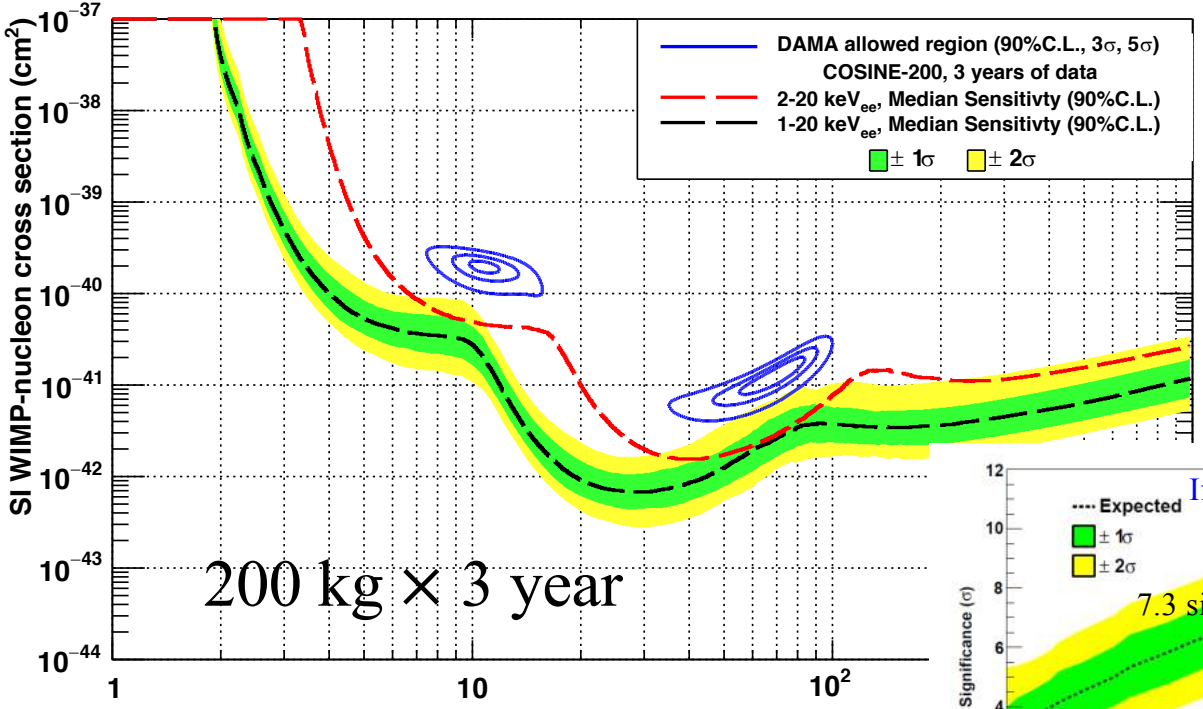
Crystal grade NaI [99.99(5) %]

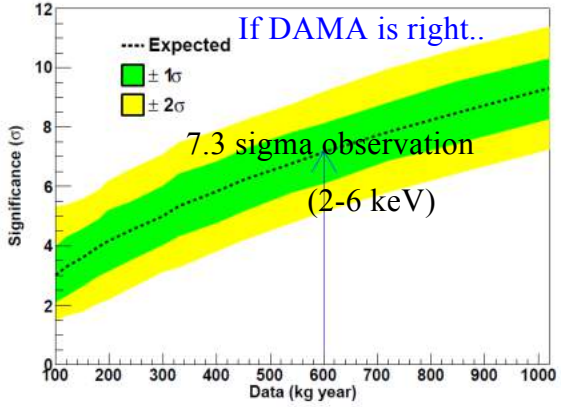
Material	Initial [ppb]	Purified [ppb]
^{39}K	45.1	6.0
^{208}Pb	3.3	0.8
^{232}Th	< 0.1	< 0.1
^{238}U	< 0.1	< 0.1

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

COSINE-200 sensitivity (Modulation)

- 1 dru background (same as DAMA/LIBRA)



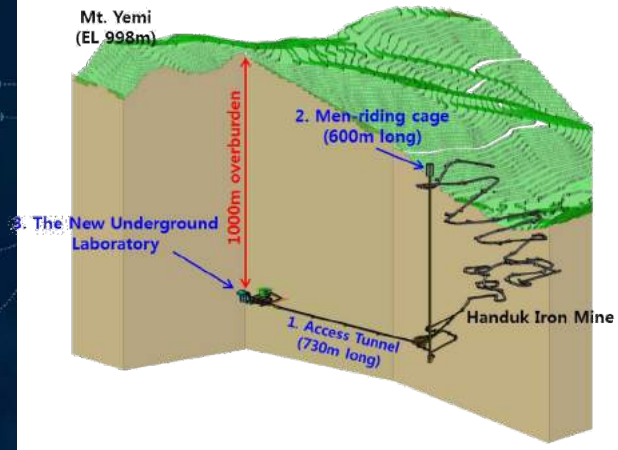


- Model independent comparison of the modulation amplitude at 2-6 keV will be performed.

Yemilab: A new underground lab in Handeok mine

- The only operating iron ore mine in Korea.
- A 600 m long 2nd shaft already constructed.
- 0.7 million tons of iron ores extracted per year

ARF is now Yemilab.

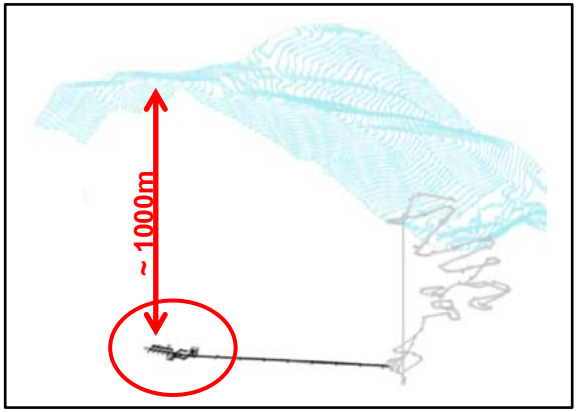


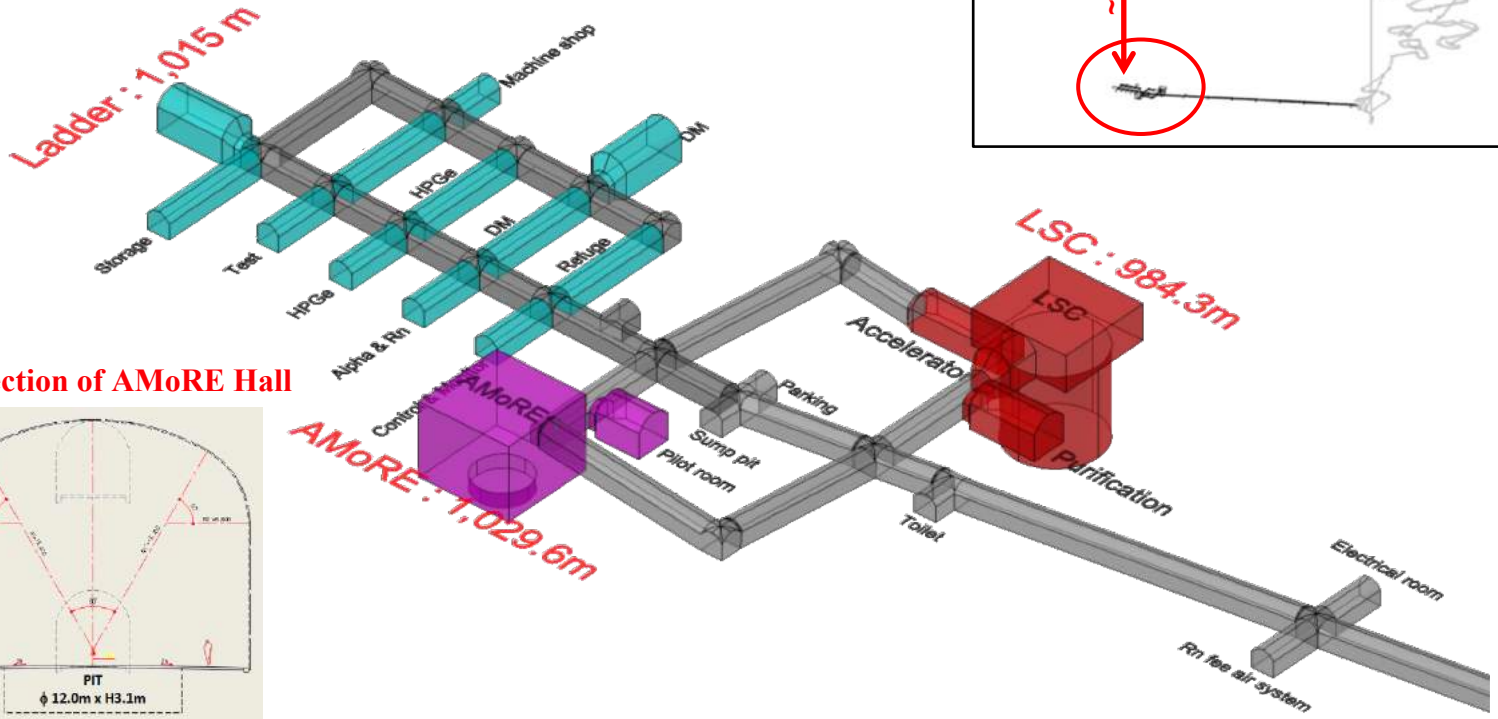
- **4 major constructions**
  - **Tunnel excavation**
  - **Shaft cage**
  - **Underground lab**
  - **Surface office/lab**

Handeok has two shafts for mining
1st shaft ~ 300 m deep
2nd shaft 600 m deep (NEW)

Yemilab: Underground Laboratories

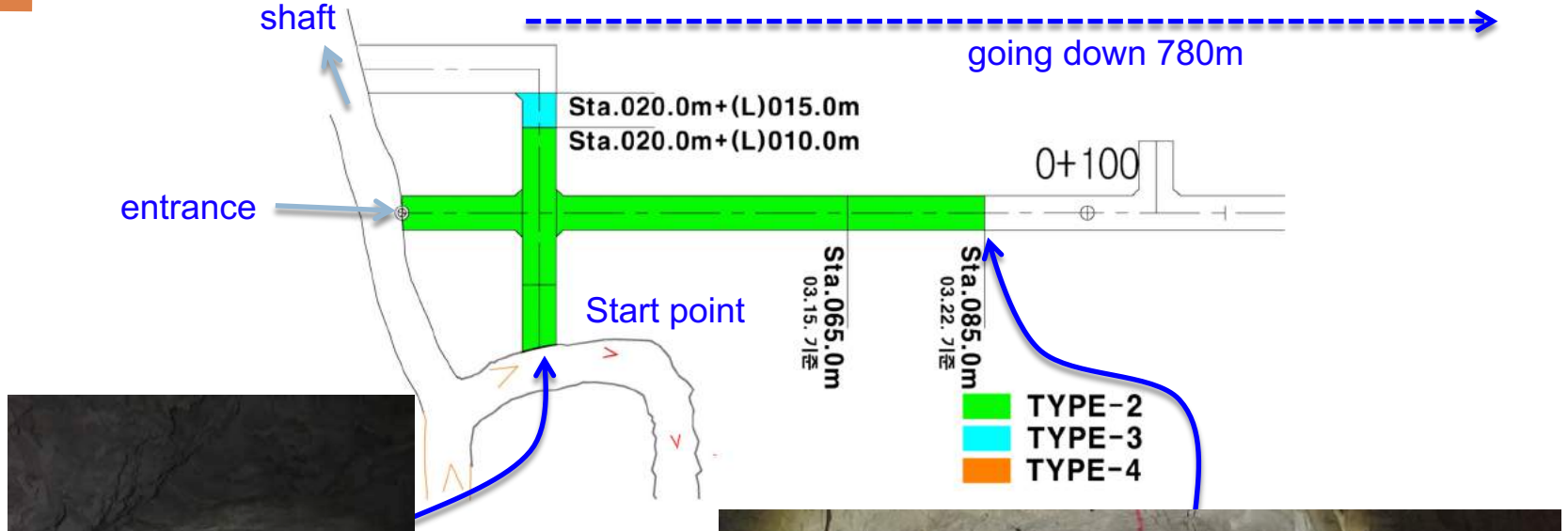
- 7 experiments with 12 spaces
- 10 utility spaces





Cross section of AMoRE Hall

Yemilab: Tunnel excavation started in March



Summary

- CUP has two major rare process experiments, AMoRE and COSINE, running in the Y2L.
- CUP has been running ultra-low background measurements facility in the Y2L and IBS HQ to screen raw materials for the detector components (i.e., crystals) from 2014.
- Background levels of AMoRE-Pilot CMO crystals measured both in RT and LT are consistent each other.
- Background levels of nine FOMOS CMO crystals measured in RT are confirmed that they meet the requirement of 10^{-3} ckky in AMoRE-I.
- AMoRE-II with 200 kg of crystals requires even lower background level of 10^{-4} ckky.
- Purification and growing of molybdate crystals at the CUP started from 2016 aiming for the AMoRE-II and in a good progress.
- COSINE-100 has 106 kg of NaI(Tl) crystals with similar or lower background levels in ^{40}K/U/Th than those of the DAMA except ^{210}Pb. It has 2-3 more background than that of the DAMA in low energy.
- COSINE's next phase is going to use ultra-pure NaI(Tl) crystals with ~200 kg mas to confirm/dispute the DAMA's annual modulation in model-independent way.
- Purification and crystal growing of NaI(Tl) started at the CUP from 2017 and in a good progress.
- A new underground lab to accommodate the AMoRE-II, called Yemilab, has started its tunnel excavation from March to be completed by the end of this year.