Copper Electroforming at Laboratorio Subterráneo de Canfranc (LSC)

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Outline

- LSC general presentation.
- CES (Copper Electroforming Service).
- Conclusions.

LSC General Presentation
External Buildings

EDIFICIO SEDE

- Headquarters & Administration
- Safety and Quality Assurance
- 23 offices
- 4 specialized laboratories
- Mechanical workshop
- Storage room
- Meeting room
- Library
- Conference & exhibition rooms
- 2 apartments

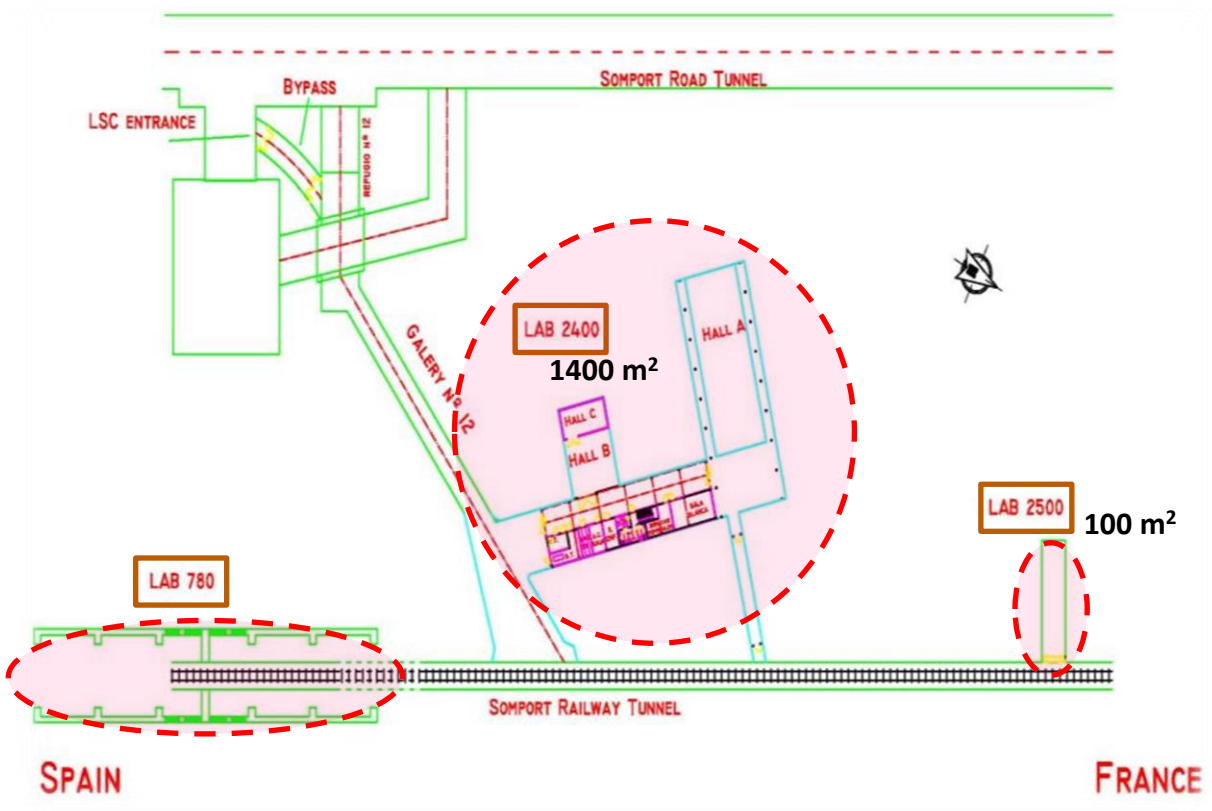
Surface: 1,821 m^2
(2,115 m^2 built)

CASA DE LOS ABETOS

- 2 Classroom
- 1 Conference room
- Exhibitions room

Surface: ~400 m^2
(460 m^2 built)

LSC General Presentation
Underground facilities



LAB2400

850 m rock overburden

2,450 meters of water equivalent (m.w.e.) depth

muon flux ~4×10^{-3} m^{-2} s^{-1}

LSC General Presentation
Underground facilities

- ULBS (Ultra Low Background Service): 7 HPGe detectors, Hall C
- CRS (Clean Room Service): underground clean room class ISO 7 (ISO 6 in a sector)
- Radon abatement system (220 m^3/ h, for NEXT, BiPo and ANAIS experiments), Hall A
- Radon detector (1 mBq m^{-3}, in progress, collaboration with G. Zuzel, Krakow), Hall A
- Mechanical workshop
- Offices

Clean Room

Radon abatement system

Mechanical workshop

LSC General Presentation
Underground facilities

Picture by J. Quiñoa

Hall A

LSC General Presentation
Current Science Program at LSC

PHYSICS

ANAIS: dark matter (NaI, annual modulation)

ArDM: dark matter (two-phase LAr TPC)

TREX: dark matter (HP Gas TPC)

NEXT: $o\nu2\beta$ decay (Enriched ^{136}Xe gas TPC)

CROSS: $o\nu2\beta$ decay (^{100}Mo bolometers)

BiPo: $o\nu2\beta$ decay (ancillary to Super-NEMO)

SuperK-Gd: material screening of Gd salts for Super - Kamiokande

CLYC-N: screening of neutrons in underground

A-KWISP: force sensor for short-range interactions

GEOPHYSICS

Geogyn: geodynamics (underground & surface) at local/ global scale

ETSEC: seismic sensors in the ET framework (characterization of Newtonian background)

BIOLOGY

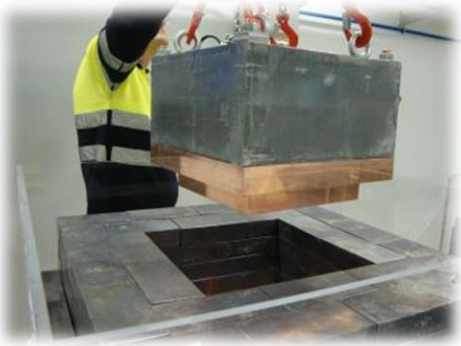
GOLLUM: life characterization deep underground

LSC General Presentation
ULBS (Ultra-Low Background Service)

7 HPGe p-type coaxial (2 kg), mounted and taking data

Hall C – LAB2400

Environmental monitoring & material screening

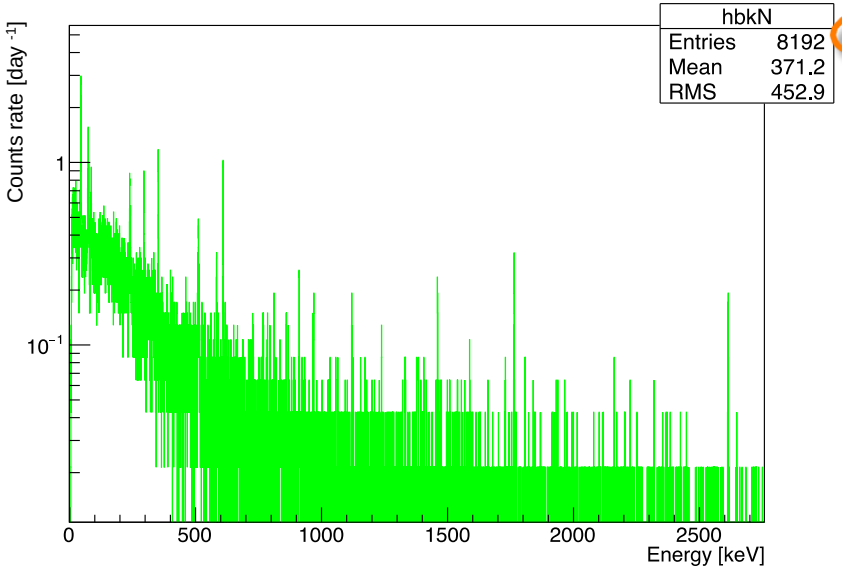


Shielding characteristics

- 5-10 cm Cu-OFHC
- 20 cm very low ^{210}Pb activity lead
- Methacrylate Rn box.
- Door
- N_2 from boil-off evaporation

LSC General Presentation
ULBS (Ultra-Low Background Service)

Typical background spectrum (GeOroel)

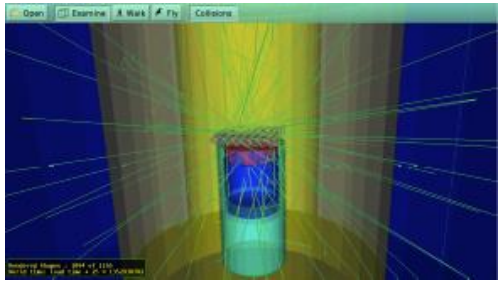


Detector Name	Diameter [mm]	Height [mm]	Weight [g]	Rel. Eff. [%]	FWHM @ 1332 keV [keV]	Cryostat Mat.
GeOroel	81.4	81.7	2230	109.0	1.85	Al
GeTobazo	81.2	81.2	2185	110.0	2.07	Al
GeAnayet	81.0	81.1	2183	109.0	1.96	Al
GeAspe	81.0	81.2	2187	108.0	1.94	Al
GeLatuca	81.0	81.2	2187	108.0	1.86	Al
Asterix	79.0	79.0	2031	95.1	2.08	Cu
Obelix	79.0	79.0	2031	92.8	2.00	Cu

Integral 40 keV -> 2700 keV day^{-1} kg^{-1} = 184 counts

^{238}U/ ^{214}Bi (609 KeV) =2.66 counts day^{-1} kg^{-1}
^{232}Th/ ^{208}Tl (583.2 Kev) = 0.57 counts day^{-1} kg^{-1}

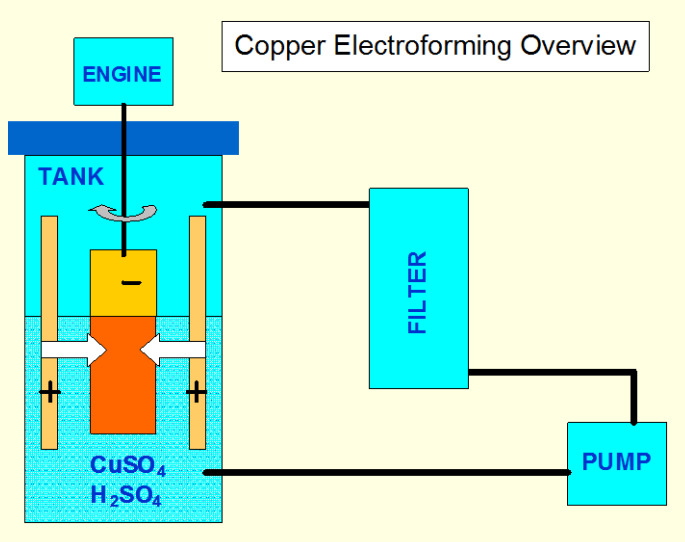
A MC simulation program, GEBIC, was developed using Geant4. Code adapted to include specific characteristics of each detector.



CES (Copper Electroforming Service)
Technique and set-up

Electroforming is a method of producing pieces by the deposition of a metal onto a mold, which is subsequently removed.

- Fixed current density direct electroplating
- High-purity commercial chemicals & ultra-pure water
- Cu-OFHC (anodes) & stainless steel 316L (cathode)



2 set-ups operative
Chemistry Lab – External Building

Technical details published at AIP Conference Proceedings 897, LRT2006

CES (Copper Electroforming Service)
Electroformed copper part

Electroformed copper endcap
(81.35 mm internal diameter, 80 mm height, 2.5 mm thickness, 627.2 g).

Process parameters: current density (3 A dm^{-2}), mold turning at 1.68 rev s^{-1} rotation speed, changing its rotation direction (forward or reverse direction) every 10 min.

- 1st mechanization treatment (only edges, to remove the dog-bone effect)
- 2nd mechanization treatment (external surface)

Electroforming process time: 90 h 30 min

Cleaning protocol (nitric acid etching and citric acid passivation)

Stored underground after the preparation

CES (Copper Electroforming Service)
Gamma spectrometry measurement

I. Bandac (LSC)

Gamma spectrometry measurement performed using a HPGe detector (GeOroel, 2Kg Ge coaxial p-type, ~100 % relative efficiency) of the electroformed copper part at ULBS (LSC).

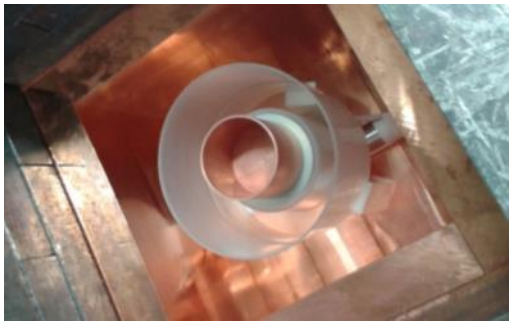
Due to the limited free space inside the sample cavity, the height of the electroformed copper part had to be reduced cutting the piece with a saw (75.4 mm and 577.8 g).

Isotope	^{234}Th	^{234m}Pa	^{235}U	^{228}Ra	^{228}Th	^{226}Ra	^{137}Cs	^{60}Co	^{58}Co	^{57}Co	^{56}Co	^{54}Mn	^{40}K
Units (mBq kg^{-1})	< 11.2	< 72	< 5.6	< 2.23	< 1.36	< 9.8	< 0.86	< 0.71	< 0.4	< 0.8	< 0.43	< 0.31	< 6.93

All the limits are 95 % C.L.

Measuring time: 50.4 days

Piece over a Marinelli container, bottom part close to the Ge detector

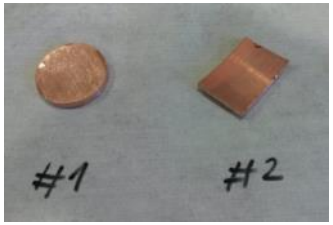


CES (Copper Electroforming Service)
Inductively Coupled Plasma Mass Spectrometry assay

S. Nisi, M.L. Di Vacri (LNGS, Italy)

Inductively-Coupled Plasma Mass Spectrometry (ICP-MS) assay carried out to analyze the Th and U contamination at the surface and bulk in copper samples

Sample preparation
- Nitric acid etching *
- Chromatographic extraction resins
- Eluted from the columns by ammonium oxalate



* Surface assay: samples etched once, ~2 g copper measured
Bulk assay: samples etched two times more, ~1 g copper measured

Sample	Description	Surface assay		Bulk assay	
		Th [ppt]	**U** [ppt]	**Th** [ppt]	**U** [ppt]
#1	OFHC copper, raw material, 15g	280	48	4.6	~1
#2	Electroformed copper obtained at CES, 12.3g	49	22	<1	<1

The error is estimated to be about 10%.

CES (Copper Electroforming Service)
Inductively Coupled Plasma Mass Spectrometry assay

I. J. Arnquist and E. W. Hoppe (PNNL, Richland)

Inductively-Coupled Plasma Mass Spectrometry (ICP-MS) assay carried out to analyze the Th and U contamination at bulk in copper samples

Sample preparation

- Nitric acid etching
  - ❖ Sample #1 totally digested by etching
  - ❖ Sample #2 cut in three pieces, 1/3 of mass etched away, remaining material completed dissolved by etching
- Pre-concentration (maximizing the analytes Th & U) and anion exchange separation (minimizing matrix)

#1 Remaining OFHC copper sample (LNGS)

#2 New electroformed copper sample

Sample	Description	Th [ppt]	+/- sd	U [ppt]	+/- sd
#1	Remaining OFHC copper	0.967	0.059	0.196	0.011
#2_LSC_01	Electroformed copper obtained at CES, three pieces	0.0426	0.0019	<0.0497	-
#2_LSC_02		0.0345	0.0037	<0.0502	-
#2_LSC_03		0.0363	0.0050	<0.0498	-

Triplicate measurements of the same sample is shown for each sample measurement as "Inst. +/- 1s".

CES (Copper Electroforming Service)
Gamma spectrometry – ICP MS comparison

OFHC Copper	ICP-MS assay					
	LNGS surface [mBq kg^{-1}]	LNGS surface [ppt]	LNGS bulk [mBq kg^{-1}]	LNGS bulk [ppt]	PNNL [mBq kg^{-1}]	PNNL [ppt]
U	0.6049	**49**	0.01234	**<1**	0.00242	**0.196**
Th	1.1382	**280**	0.01869	**4.6**	0.00393	**0.967**

EF Copper	Gamma spectrometry HPGe		ICP-MS assay					
	LSC GeOroel [mBq kg^{-1}]	LSC GeOroel [ppt]	LNGS surface [mBq kg^{-1}]	LNGS surface [ppt]	LNGS bulk [mBq kg^{-1}]	LNGS bulk [ppt]	PNNL [mBq kg^{-1}]	PNNL [ppt]
U	**< 11.2 (^{234}Th)**	< 907.2	0.2716	**22**	< 0.012	**<1**	< 0.000614	**<0.0498**
Th	**< 2.23 (^{228}Ra)**	< 548.5	0.1992	**49**	< 0.0406	**<1**	0.000147	**0.0363**

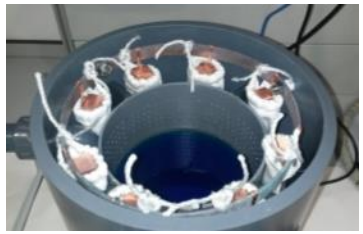
1 Bq ^{238}U/kg = 81x10^{-9} g/g (ppb)
1 Bq ^{232}Th/kg = 246x10^{-9} g/g (ppb)

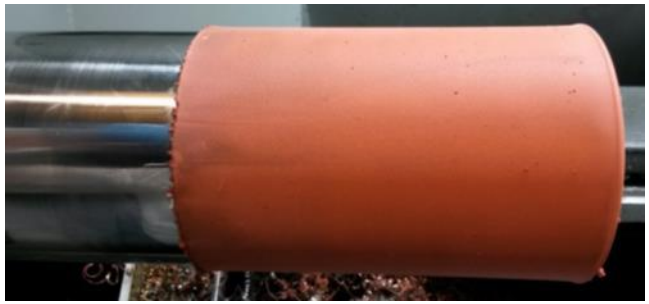
CES (Copper Electroforming Service)
LNGS & LSC Collaboration

Electroformed copper cylinder using Cu-ETP (UNI 5649-71) bars from LNGS (81.35 mm diameter, 118 mm height, 2.3 mm thickness, 547.6 g).

Process parameters: current density (3 A dm^{-2}), mold turning at 1.68 rev s^{-1} rotation speed, changing its rotation direction (forward or reverse direction) every 10 min.

- Two mechanization treatments (only edges, to remove the dog-bone effect)
- None final surface mechanization treatment





Electroforming process time: 75 h

None cleaning protocol

Stored underground after the preparation

CES (Copper Electroforming Service)
LNGS & LSC Collaboration

S. Nisi, M.L. Di Vacri (LNGS, Italy)

Inductively-Coupled Plasma Mass Spectrometry (ICP-MS) assay carried out for a semi-quantitative analysis at bulk in copper samples

Sample preparation

- Cleaning protocol (5% citric acid)
- Nitric acid etching (two steps)
  ~3.6 – 3.8 g of sample measured from the second etching

Reference Standard solution 10 ppb (Li, Ce, Y, Tl) added to a portion of the sample solution to evaluate the instrumental response

Work in progress

	Starting copper (Cu ETP from LNGS)	Electroformed copper obtained at LSC	Removal efficiency
	[ng g^{-1}]	[ng g^{-1}]	[%]
Fe	13 x 10^3	< 3 x 10^3	> 76.9
Co	1.6 x 10^3	< 1	> 99.9
Ag	1.0 x 10^3	240	76.0
Sn	19 x 10^3	< 5	> 99.9
Sb	1.9 x 10^3	< 5	> 99.7
Nd	< 5	130	
Bi	180	< 5	> 97.2
Pb	49 x 10^3	< 50	> 99.9

The uncertainty of results is estimated to be about 30% of the given values.

"ICP-MS measurements of ultra-low level radioactivity in solid material and comparison with γ– spectrometry within the CUPID experiment" M.L. Di Vacri (ICRM-LLRMT meeting, 2016).

CES (Copper Electroforming Service)
Requests & Collaborations

From 2014 performed electroformed copper pieces: ANAIS, LNGS Collaboration…

- ANAIS Experiment: 28 electroformed copper parts for 14 PMT encapsulation.

EF copper endcap

NaI(Tl) module assembly in the LSC Clean Room

Current configuration of ANAIS experiment

(Pictures courtesy of ANAIS Collaboration)

- LNGS Collaboration (Italy): study of the copper purification by the electroforming technique.

Work in progress

- Jagiellonian Univ. Collaboration (Poland): study of the residual $^{210}Pb/^{210}Po$ bulk content, surface radiopurity and searches for appropriate surface cleaning protocols of electroformed copper.

Work in progress

Conclusions

- The CES facilities allow to carry out R&D activities and give support to experiments working at LSC.
- Current cleaning protocol is not sufficient to clean surface contamination from the electroformed copper pieces.
- The comparison ICP MS - gamma spectrometric measurements demonstrates that the techniques are in agreement and complementary.
- The electroforming process performed at LSC resulted to be efficient for copper purification for several elements of interest.
- CES is being used to carry out several R&D activities in collaboration with other Laboratories and research.

Thank you!