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

CASA DE LOS ABETOS
• 2 Classroom
• 1 Conference room
• Exhibitions room

Surface: 1,821 m² (2,115 m² built)
Surface: ~400 m² (460 m² built)
LAB 2400

850 m rock overburden

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

Muon flux \(~4\times10^{-3}\) 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
# 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**: $0\nu\beta\beta$ decay (Enriched $^{136}$Xe gas TPC)

**CROSS**: $0\nu\beta\beta$ decay ($^{100}$Mo bolometers)

**BiPo**: $0\nu\beta\beta$ 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
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
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)

Technical details published at AIP Conference Proceedings 897, LRT2006
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
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).

<table>
<thead>
<tr>
<th>Isotope</th>
<th>$^{234}$Th</th>
<th>$^{234m}$Pa</th>
<th>$^{235}$U</th>
<th>$^{228}$Ra</th>
<th>$^{228}$Th</th>
<th>$^{226}$Ra</th>
<th>$^{137}$Cs</th>
<th>$^{60}$Co</th>
<th>$^{58}$Co</th>
<th>$^{57}$Co</th>
<th>$^{58}$Co</th>
<th>$^{54}$Mn</th>
<th>$^{40}$K</th>
</tr>
</thead>
<tbody>
<tr>
<td>Units</td>
<td>&lt; 11.2</td>
<td>&lt; 72</td>
<td>&lt; 5.6</td>
<td>&lt; 2.23</td>
<td>&lt; 1.36</td>
<td>&lt; 9.8</td>
<td>&lt; 0.86</td>
<td>&lt; 0.4</td>
<td>&lt; 0.8</td>
<td>&lt; 0.43</td>
<td>&lt; 0.31</td>
<td>&lt; 6.93</td>
<td></td>
</tr>
<tr>
<td>(mBq kg$^{-1}$)</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

All the limits are 95 % C.L.

Measuring time: 50.4 days

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

<table>
<thead>
<tr>
<th>Sample</th>
<th>Description</th>
<th>Surface assay</th>
<th>Bulk assay</th>
</tr>
</thead>
<tbody>
<tr>
<td>#1</td>
<td>OFHC copper, raw material, 15g</td>
<td>280</td>
<td>48</td>
</tr>
<tr>
<td>#2</td>
<td>Electroformed copper obtained at CES, 12.3g</td>
<td>49</td>
<td>22</td>
</tr>
</tbody>
</table>

The error is estimated to be about 10%.
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)

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

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

<table>
<thead>
<tr>
<th></th>
<th>LSC GeOroel [mBq kg(^{-1})]</th>
<th>LSC GeOroel [ppt]</th>
<th>LSC GeOroel [mBq kg(^{-1})]</th>
<th>LSC GeOroel [ppt]</th>
<th>LSC GeOroel [mBq kg(^{-1})]</th>
<th>LSC GeOroel [ppt]</th>
<th>PNNL [mBq kg(^{-1})]</th>
<th>PNNL [ppt]</th>
</tr>
</thead>
<tbody>
<tr>
<td>U</td>
<td>0.6049</td>
<td>49</td>
<td>0.01234</td>
<td>-1</td>
<td>0.00242</td>
<td>0.196</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Th</td>
<td>1.1382</td>
<td>280</td>
<td>0.01869</td>
<td>4.6</td>
<td>0.00393</td>
<td>0.967</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

#### EF Copper

<table>
<thead>
<tr>
<th></th>
<th>LSC GeOroel [mBq kg(^{-1})]</th>
<th>LSC GeOroel [ppt]</th>
<th>LSC GeOroel [mBq kg(^{-1})]</th>
<th>LSC GeOroel [ppt]</th>
<th>LSC GeOroel [mBq kg(^{-1})]</th>
<th>LSC GeOroel [ppt]</th>
<th>PNNL [mBq kg(^{-1})]</th>
<th>PNNL [ppt]</th>
</tr>
</thead>
<tbody>
<tr>
<td>U</td>
<td>&lt; 11.2 (^{234})Th</td>
<td>&lt; 907.2</td>
<td>0.2716</td>
<td>22</td>
<td>&lt; 0.012</td>
<td>&lt;1</td>
<td>&lt; 0.000614</td>
<td>&lt;0.0498</td>
</tr>
<tr>
<td>Th</td>
<td>&lt; 2.23 (^{228})Ra</td>
<td>&lt; 548.5</td>
<td>0.1992</td>
<td>49</td>
<td>&lt; 0.0406</td>
<td>&lt;1</td>
<td>0.000147</td>
<td>0.0363</td>
</tr>
</tbody>
</table>

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

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).

![Work in progress](image)
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
  - Nal(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 $^{210}\text{Pb}/^{210}\text{Po}$ 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!