# The MAJORANA DEMONSTRATOR Assay Program and Capabilities Necessary to Support a Future 1 Tonne Scale Experiemnt

E.W. Hoppe

Pacific Northwest
NATIONAL LABORATORY

*Proudly Operated by* Battelle *Since 1965*

# Top Level Table for Materials-MAJORANA

| Material | Part of Demonstrator | Decay Chain | Achieved Assay | | Reference |
|---|---|---|---|---|---|
| | | | [μBq/kg] | [c/ROI/t/y] | |
| EFCu | Inner Cu Shield, Cryostat, Coldplate, Thermal Shield, Detector Mounts | Th | 0.06 | 0.15 | [1] |
| | | U | 0.17 | 0.08 | |
| OFHC | Outer Copper Shield (O.Cut) | Th | 1.10 | 0.24 | [2] |
| | | U | 1.25 | 0.04 | |
| Pb | Lead Shield | Th | <32 | <1.5 | [3] |
| | | U | <112 | <1.2 | |
| PTFE | Detector Supports | Th | 0.1 ± 0.01 | 0.01 | [4,5] |
| | | U | <5 | <0.01 | |
| PEEK | Cross Arm Support | Th | <1600 | <0.01 | [4,5] |
| | | U | <63000 | <0.01 | |
| Vespel | Cold Plate Support | Th | <12 | <0.01 | [4,5] |
| | | U | <1050 | <0.01 | |
| Parylene | Cu coating, Cryostat seals | Th | 2150 | 0.16 | [6] |
| | | U | 3110 | 0.05 | |
| Silica / Au, Epoxy | Front-End Electronics | Th | 6530 | 0.32 | [7] |
| | | U | 10570 | 0.28 | |
| Cu Wire + PFA | Signal /HV Cable and Connectors | Th | 2.2 | 0.01 | [8] |
| | | U | 145 | 0.14 | |
| Stainless Steel | Service Body | Th | 13000 | <0.04 | [9] |
| | | U | <5000 | <0.03 | |


[1] "Determination of Method Detection Limits for Trace 232-Thorium and 238-Uranium in Copper using Ion Exchange and ICPMS-January 2014", [M-TECHDOCPhys-2014-72].

[2] Commercial Copper Assay Report[M-TECHDOCDET-2012-149]

[3] Results of Lead Assay by GDMS; final [M-TECHDOCDET-2012-146]

[4] Final report on assay of plastic for MAJORANA DEMONSTRATOR [M-TECHDOCDET-2011-137].

[5] Report on NAA performed at HFIR (ORNL) for samples of TE-6742 for the MAJORANA DEMONSTRATOR [M-TECHDOCDET-2011-128].

[6] Cable Assay [M-TECHDOCDET-2011-124].

[7] ICP-MS of Low Mass Front-End Board; final [M-TECHDOCDET-2013-157]

[8] ICP-MS Assay of MJD Copper Cables for Uranium & Thorium - December 2013; [M-TECHDOCMJTEST-2014-027].

[9] Service Body Assay Results; final [M-TECHDOCDET-2012-143]

# Top Level Table for Materials-MAJORANA

| Material | Part of Demonstrator | Decay Chain | Achieved Assay | | Method |
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# Copper Electroforming (EFCu)



- Production of EFCu
  - 10 baths at the TCR in SURF
    - 1 bath now decommissioned Nov 2013
  - 7 baths (out of 14) in PNNL underground
    - completing production for MJD in Jan 2014

*Inspection of copper being electroformed at the TCR in SURF (above) and the shallow underground laboratory at PNNL (left)*

MJD target of 0.3 µBq for both $^{232}$Th and $^{238}$U /kg Cu to meet equivalent of 1 count/ROI/t/y

# Selecting Assay Techniques that Support Need

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- Quantitative assessment of background benefit from a variety of assay methods

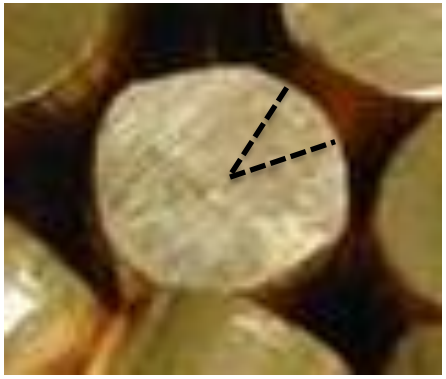
Sample: 24.23 kg OFHC Cu nuggets, 2009

**GeMPI-2 at Gran Sasso (4 month count or 8 kg-yr):**

- $^{40}$K 190 ± 60 μBq/kg
- $^{228}$Th 30 ± 10 μBq/kg
- $^{226}$Ra 20 ± 10 μBq/kg

(thanks to M. Laubenstein)

Sample 0.01 kg OFHC Cu nugget, 2009



**ICP-MS at PNNL (1 week):**

- $^{40}$K <500 μBq/kg
- $^{232}$Th 1.3 ± 0.6 μBq/kg
- $^{238}$U <30 μBq/kg

**Combined results provides most useful information (2009):**

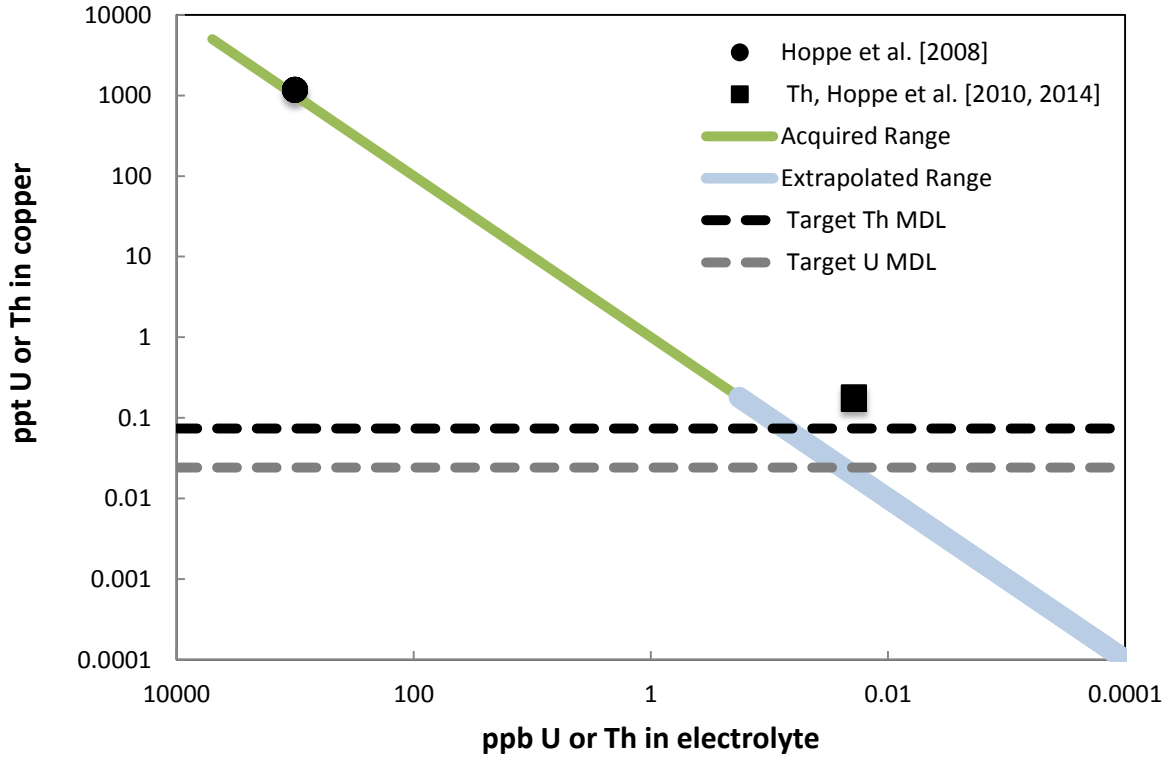
- $^{40}$K 190 ± 60 μBq/kg
- $^{232}$Th 1.3 ± 0.6 μBq/kg
- $^{238}$U 20 ± 10 μBq/kg

Current PNNL ICP-MS now (Jan 2013) sensitive to lower levels:

- $^{232}$Th ~0.7 μBq/kg
- $^{238}$U ~1.3 μBq/kg

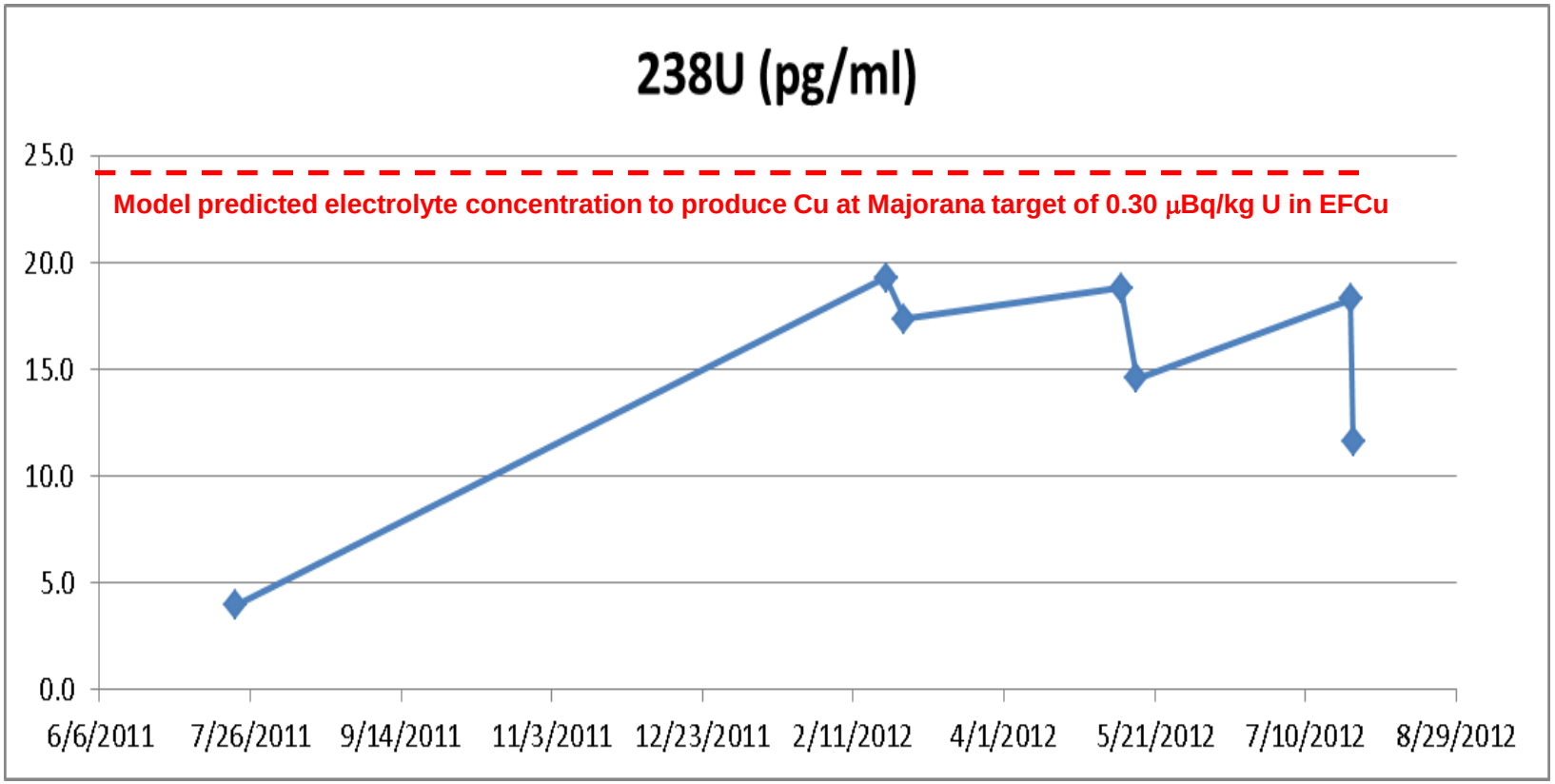
# Indirect Copper Purity Assay

In the absence of direct assays of copper with adequate sensitivity for U and Th, a simple model is produced to predict the concentration of these impurities based on the electrolyte assays. Based on this, MJD begins production electroforming in Sept 2010

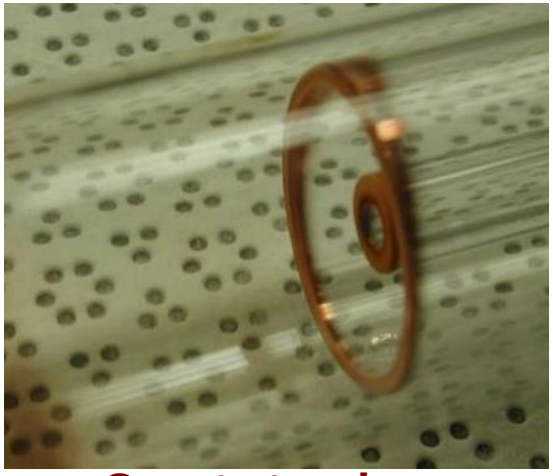


# Indirect Copper Purity Assay

Example of electrolyte replacement when contaminant levels reach the upper operating range Bath 3 TCR, first production run. PNNL has conducted over 300 electrolyte assays (as of Sept 2013) to track quality of the electroforming baths

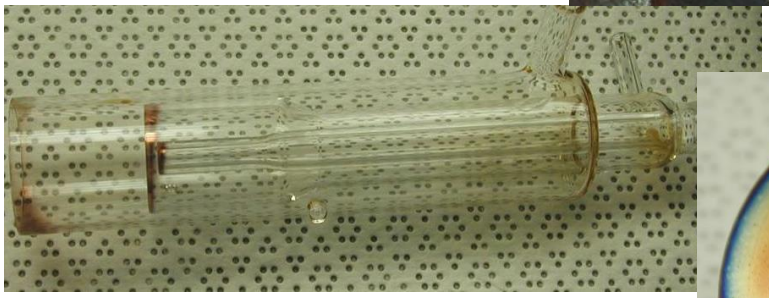


# Too much matrix (copper) issues during ICPMS analysis

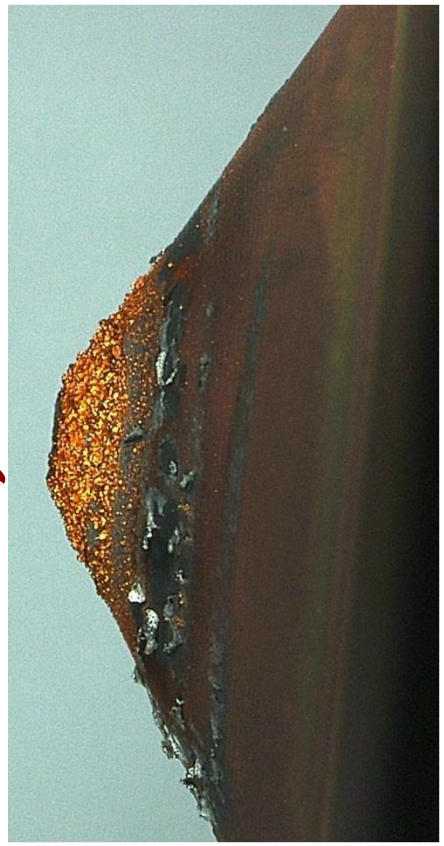


Quartz torch

Ni cones



X-lens



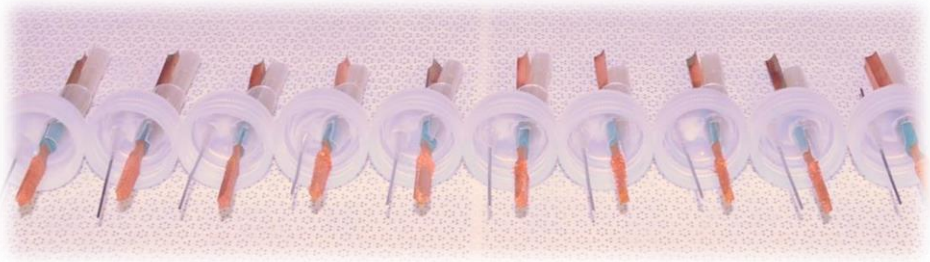
ICPMS does not possess adequate dynamic range to simply dissolve the sample prior to analysis and expect sensitivity desired for current experiments

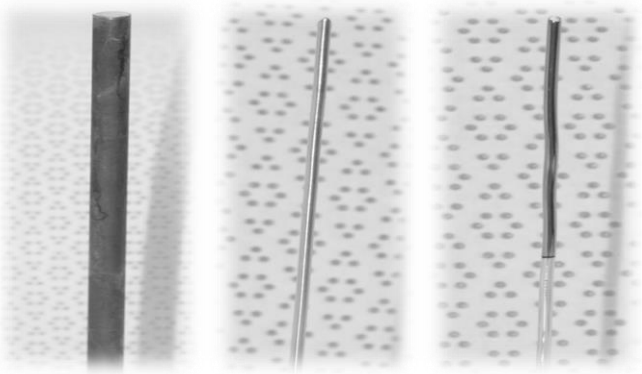
# Copper Assay Development



- Completed the development of two methods at PNNL in 2013, electrochemical and ion exchange sample preparation, for trace assay of $^{232}$Th and $^{238}$U in copper
- Both have an MJD target of 0.3 μBq/kg Cu
  - Method Detection Limits for the electrochemical sample preparation (using PNNL produced Ir electrodes) followed by ICPMS have been determined on multiple occasions by one analyst with the best obtained being 0.10 μBq for both $^{232}$Th and $^{238}$U/kg Cu

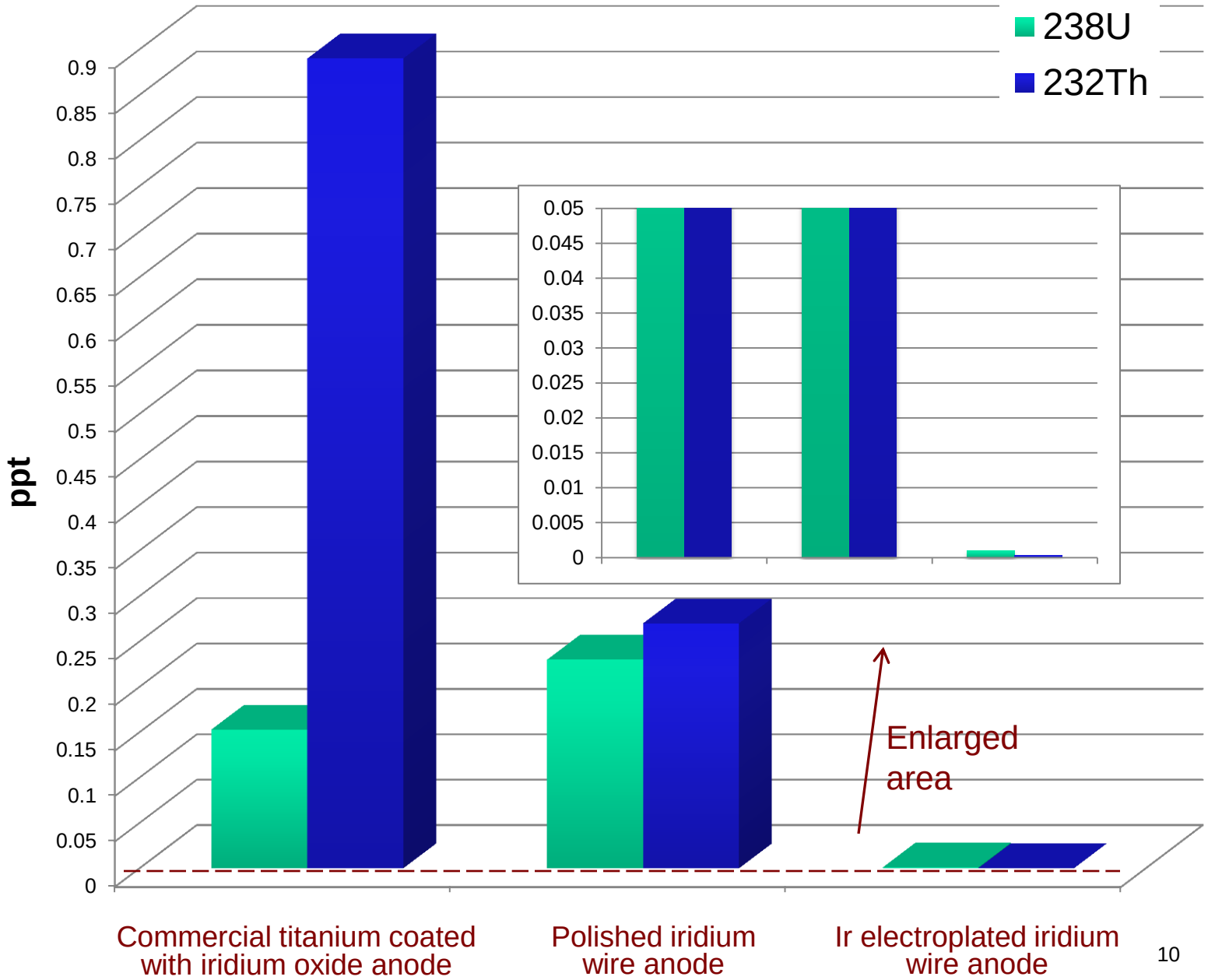
*Preparation of copper samples using the electrochemical method (above) and electrochemical electrodes (below)*





# U, Th background contributions-Electrochemical Sample Preparation

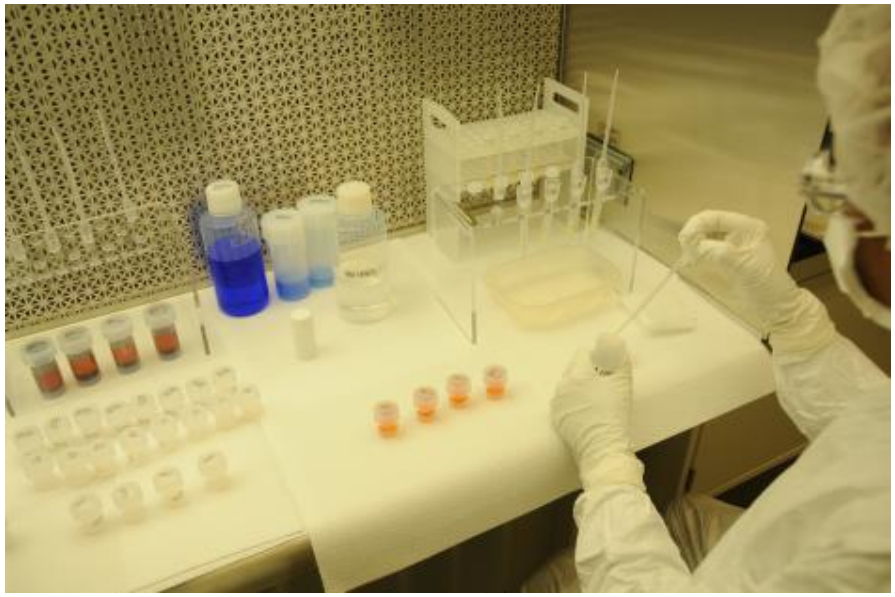
Background contributions based on a typical ~70h run time in 15ml of solution



# PNNL Activity: Copper Assay Development



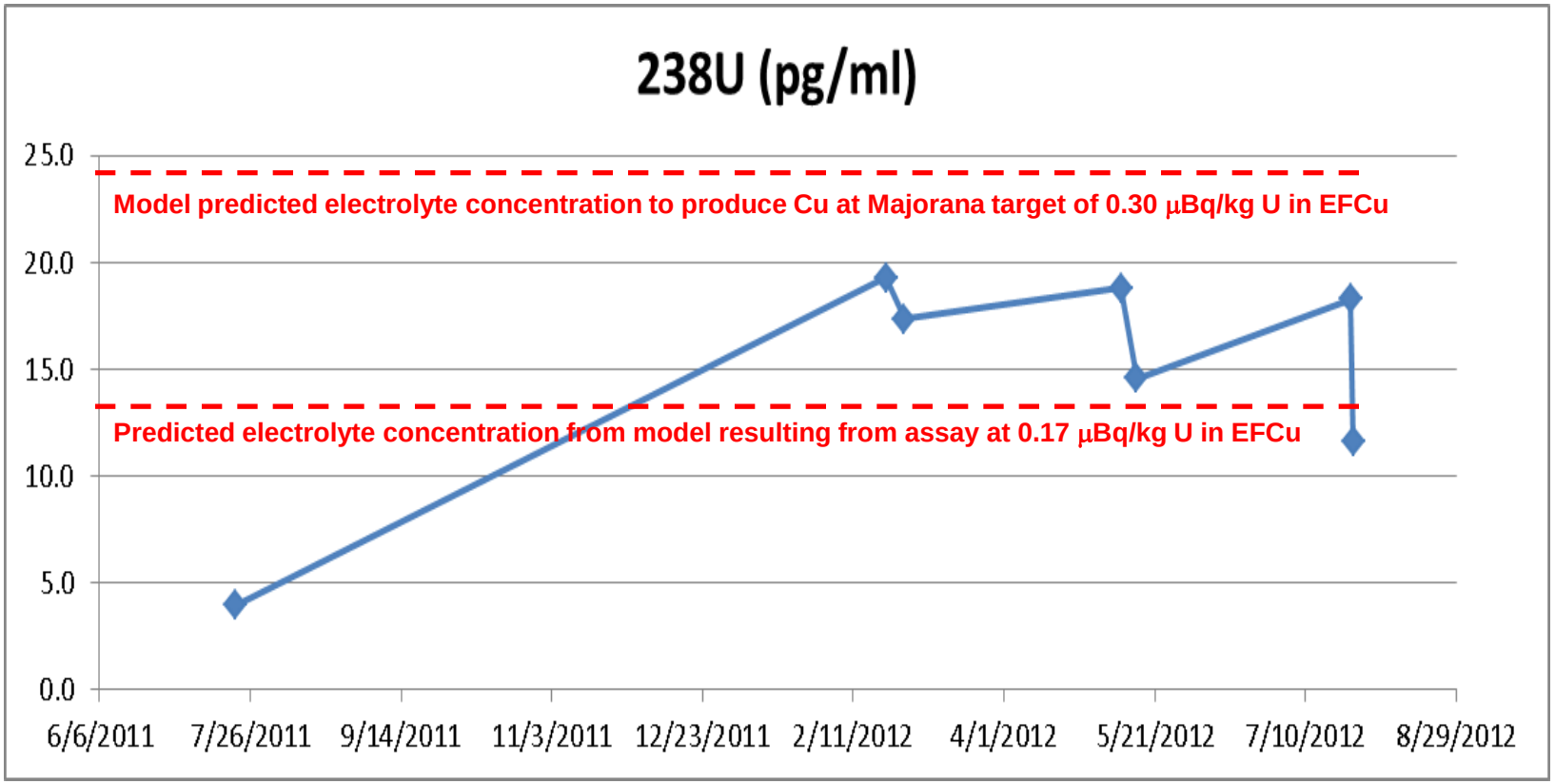
- Method Detection Limits for the ion exchange sample preparation followed by ICPMS have been determined on multiple occasions by two analysts with the best obtained being 0.045 µBq $^{232}$Th and 0.098 µBq $^{238}$U/kg Cu
- Better sensitivity and sample throughput make the ion exchange method the primary analytical method. The electrochemical method will be used as a confirming method if deemed necessary.



*Preparation of copper samples using ion exchange methods in a laminar flow hood inside a class 10000 clean room*

# Indirect Copper Purity Assay-Validation of the model

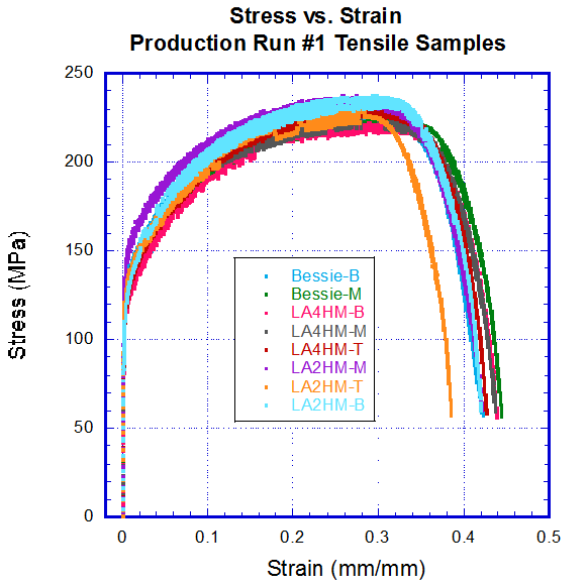
Example of electrolyte replacement when contaminant levels reach the upper operating range Bath 3 TCR, first production run. PNNL has conducted over 300 electrolyte assays (as of Sept 2013) to track quality of the electroforming baths



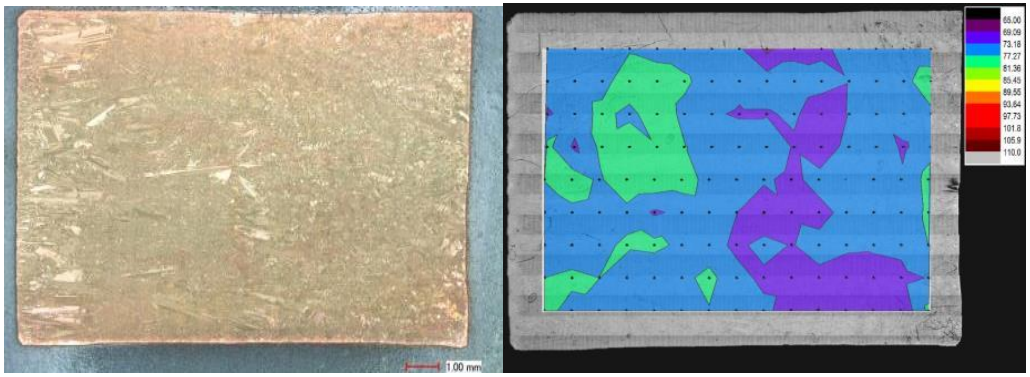
# Other Assays to Consider: Evaluation of Physical Properties



- Physical Property evaluation is comprehensive and performed for every production run
  - Important even if not producing your own material. Example, commercial OFHC copper mechanical strength can vary significantly
  - Multiple samples from various locations
  - Hardness mapping of every electroformed cross-section
  - Tensile strength
  - Crystal structure evaluation
  - Cross comparison of data to assure consistency of material



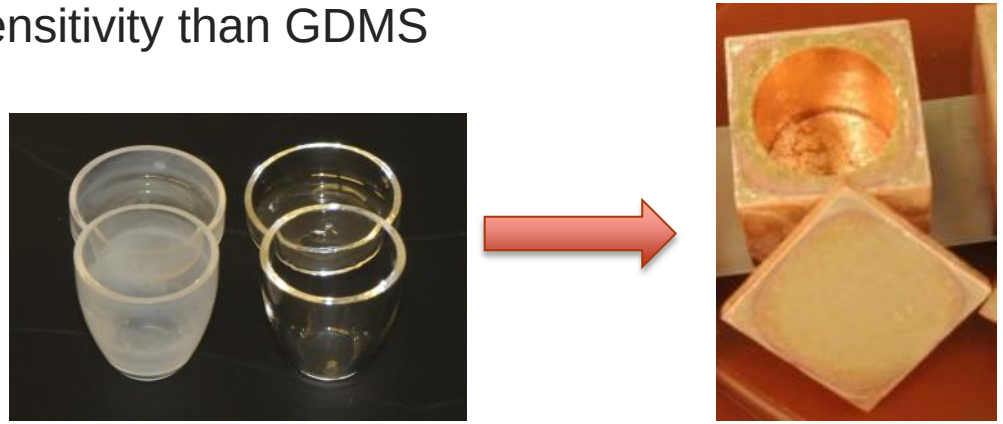

*Above-Overlay of multiple tensile strength determinations on EFCu. Below-Crystal structure and hardness mapping overlay*



# Materials Assay



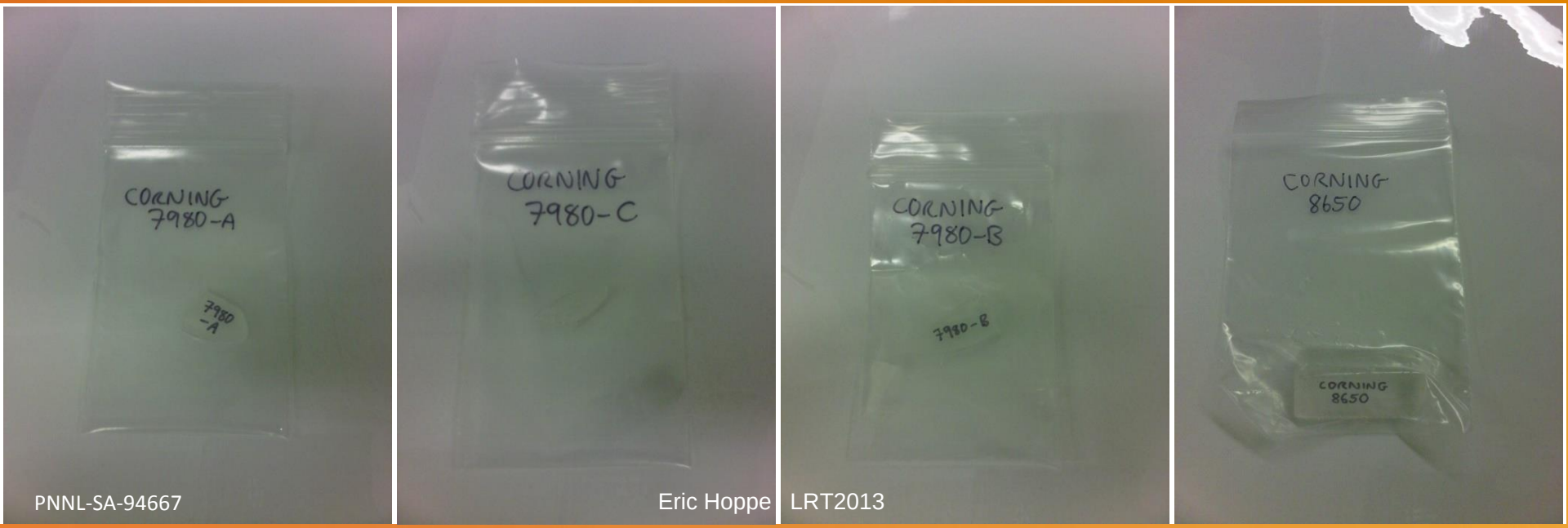
- ICP-MS assay on a wide variety of materials at PNNL
  - Assay cables and cable components, verification of new cable vendor performance
  - Polymer assays
    - Both of these use combustion in quartz furnace, microwave digestion (if necessary) followed by ion exchange
  - Continued  assay of electrolyte from production baths to determine that Th and U levels maintained better than quality target levels used for inferred assays of Cu
  - Now performing assay of Pb using modification of copper ion exchange method to achieve greater sensitivity than GDMS



*Left-Assay of polymers was challenged by the degradation of crucibles used for ashing Right-high purity copper is now used as the material of choice for ashing polymers*

# Analysis of High Purity Fused Silica and Front End Electronics

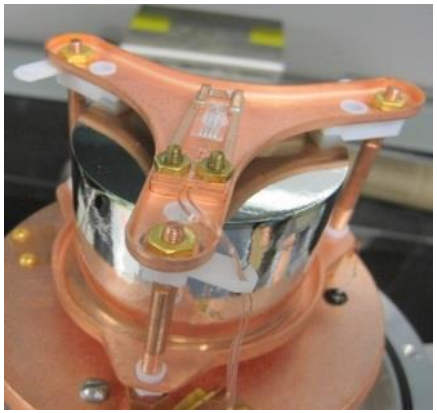
- Dissolve fused silica using HF in microwave digestor
- May also digest entire front end electronics board (FET, resistor, gold traces, etc. on fused silica wafer)
- Requires change-out of some ICP-MS components prior to HF use
- Assay detection limits for Th and U is μBq/kg, although materials not found that clean to date



# Gamma Assay at LBC



- The vacuum service body was assayed in the low background counting (LBC) lab at the Kimballton Underground Research Facility (KURF). Presented in this report are activities from detected isotopes, all of which are nuclides of naturally occurring radioactive materials (NORM) with the exception of $^{60}$Co. It was found that $^{60}$Co had the highest activity in the detector region, measuring 18.2 ± 1.0 mBq/kg.



# NAA performed at HFIR (ORNL) of polymer samples



- Polymer samples were analyzed via NAA analysis. A combination of irradiation at HFIR reactor, above ground fast counting at ORNL, and underground low background counting were used.
- Sample preparation remains a critical issue-contamination during initial sample preparation limited the method sensitivity, especially for Uranium detection.
- Upper limit of 1.15 pg $^{238}U/g$ was set. $^{232}Th$ was detected in the samples at a concentration of 0.025±0.002 pg/g.

# Follow on NAA performed at HFIR (ORNL)

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| Plastic | PTFE (TE-6742) | Vespel | PEEK | Kynar |
|---|---|---|---|---|
| $^{238}$U | <0.398 pg/g | <0.398 pg/g | <5.1 ng/g | 1110 ng/g |
| $^{232}$Th | 0.025±0.002 pg/g | <2.9 pg/g | <0.4 ng/g | 1.93 pg/g |

# GDMS on samples of Pb



- Advantage is that many elements can be assayed at a time
- Samples needs to be electrically conductive (or made that way)
- Still 1-2 orders magnitude greater sensitivity desired

National Research Council Canada
**Institute for National Measurement Standards**
Analyst: B. Methven/K. Swider
**Glow Discharge Mass Spectrometric Report**
June 20, 2012
To: Yuri Efremenko
Oak Ridge National Laboratory
NO. 32723
**ISO/IEC 17025**
**Sullivan Pb, results in ng/g**

| | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|
| 1 H | 2 Li <0.2 | 3 Be <0.08 | 4 B <0.1 | 5 C 180 | 6 N 3 | 7 O 200 | 8 F <0.5 | 9 Na 26 | 0 Mg <0.2 |
| 1 Al <0.2 | 2 Si 94 | 3 P <0.2 | 4 S 8 | 5 Cl 18 | 6 K 4 | 7 Ca <2 | 8 Sc <0.1 | 9 Ti <0.05 | 0 V <0.04 |
| 1 Cr <0.2 | 2 Mn <0.1 | 3 Fe <0.1 | 4 Co <0.09 | 5 Ni 89 | 6 Cu 100 | 7 Zn <2 | 8 Ga <0.4 | 9 Ge <0.9 | 0 As <0.2 |
| 1 Se 110 | 2 Br <0.8 | 3 Rb <0.1 | 4 Sr <0.1 | 5 Y <0.09 | 6 Zr <0.2 | 7 Nb <0.09 | 8 Mo <0.5 | 9 Ru | 0 Rh |
| 1 Pd | 2 Ag 12000 | 3 Cd 280 | 4 In <0.3 | 5 Sn <1 | 6 Sb 10 | 7 Te <0.5 | 8 I <0.1 | 9 Cs <0.09 | 0 Ba <0.2 |
| 1 La <0.06 | 2 Ce <0.08 | 3 Pr | 4 Nd | 5 Sm | 6 Eu | 7 Gd | 8 Tb | 9 Dy | 0 Ho |
| 1 Er | 2 Tm | 3 Yb | 4 Lu | 5 Hf <0.3 | 6 Ta | 7 W <0.4 | 8 Re | 9 Os | 0 Ir |
| 1 Pt <0.9 | 2 Au <30 | 3 Hg <5 | 4 Tl <140 | 5 Pb Matrix | 6 Bi 400 | 7 Th <0.01 | 8 U <0.01 | | |

Note: Due to the semi-quantitative capabilities of GD-MS, the determined mass fractions of impurities may lie in the range of one-half- to two-fold reported values except for C, N and O, for which the range is one-fifth- to five-fold.