Ultra-High-Purity Copper Technology Update

Craig Aalseth

Pacific Northwest National Laboratory

Majorana Update to GERDA
June 10 - 14, 2007, IRMM, Belgium

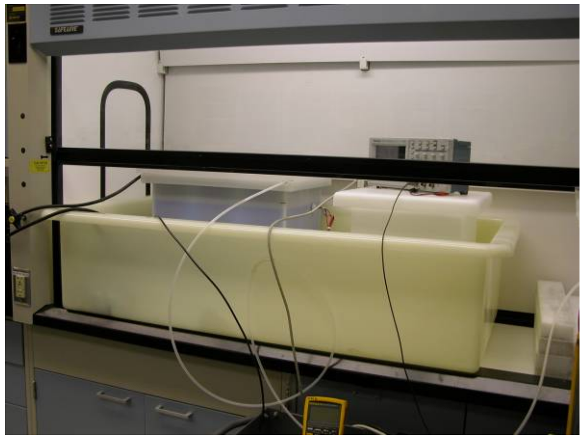
Topics

- Copper Electroforming Status
  - PNNL above-ground labs
  - LANL underground work at WIPP
- Electroformed Copper Purity
  - ^{232}Th Assay
  - ^{228}Th Tracer Study
- Electron-Beam Welding
- Copper Emissivity Measurements

Copper Electroforming Status

PNNL Chemistry Prototyping Labs 101/102

- UHP electroformed copper R&D work
  - Bath chemistry
  - Waveforms
  - Material property parametric studies
- Large-component cleaning & passivation

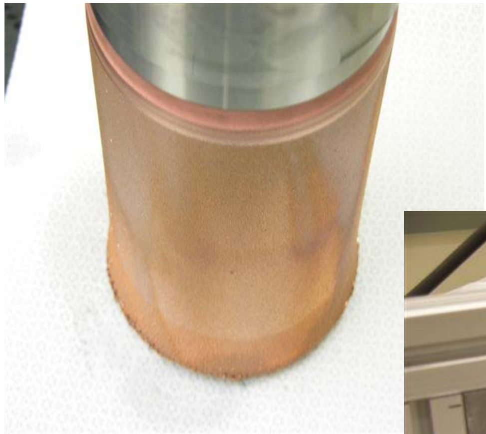


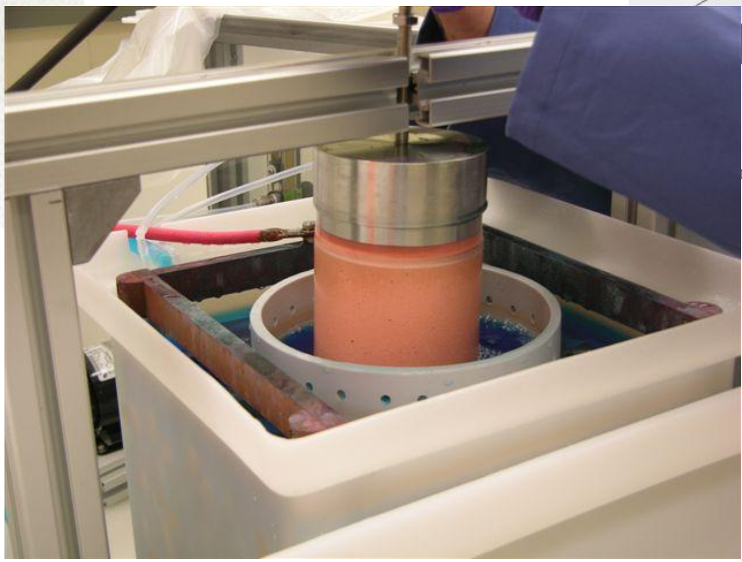
PNNL Chemistry Production Lab – 8C

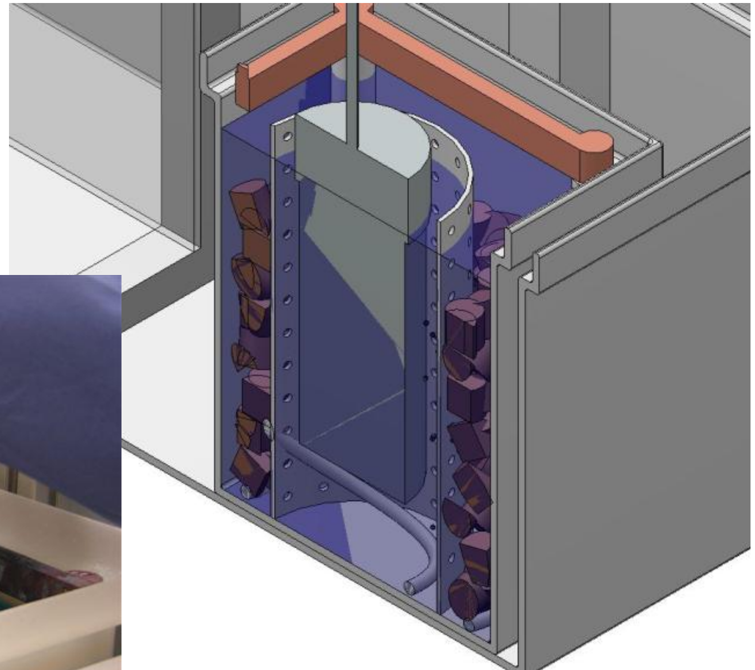
- 8 Plating Baths Constructed
  - 2 currently plating
  - 6 Prepared, currently in leaching / clean up stage
  - Some procurement / regulatory delays – now resolved
- One 12-channel programmable power supply
- Three other specialty programmable power supplies
- N2 cover gas & sparge
- Heat exchanger for temp control
- Continuous non-contact monitoring of conductivity
- Positive pressure room / HEPA filtered air
  - Class <2000 (~1300) – with some difficulty

Production Copper Electroforming

- Plating for weeks without machining, Cu ~cm thick
- Plating still slower than desired, 0.002-0.005”/day
- Developing better recipes which may improve plating rate
- ^{228}Th tracer studies will determine purity limitations vs. plating rate







Underground Electroforming Demonstration at WIPP

- **LANL demonstration of underground electroforming**
- **Test piece of 660 grams plated in 9 days**
- **Supports**
  - **ES&H process (DOE site)**
  - **Test technology transfer to UG location**
  - **Short-lived activation products measurements**
  - **Plan includes controlled surface exposure, then UG counting in LANL WIPP UG measurement facility**

Copper Production Summary

- **Underground test setup now operational at WIPP**
- **R&D space now separate from production lab at PNNL – more capacity, better cleanliness**
- **8 production baths now built, 2 operational**
- **Schedule of parts production planned for summer and fall 2007**
  - **GERDA test pieces are in this schedule**

Electroformed Copper Purity

^{232}Th Assay

^{228}Th Tracer Study

Majorana Update to GERDA
June 10 - 14, 2007, IRMM, Belgium

Status of Copper Assay and Other Measurements

- **Goals**
  - Mass Spectrometry of ^{232}Th in copper at Majorana target levels (radiochemistry + ICP-MS)
  - Continue study and control of copper electroforming process
- **New Results**
  - New positive measurement of ^{232}Th in Cu
  - Identification (and resolution) of bath purity issue
  - Additional R&D
    - ^{228}Th tracer study

Results from Copper Samples using Ion Exchange Sample Processing into ICP-MS

	Ave of µBq ^{232}Th/kg in Blanks	µBq ^{232}Th/kg of Starting Anode Cu	µBq ^{232}Th/kg of Electroformed Cu
Column 1	1.0	1.7	1.6
Column 2	0.5	1.6	1.2
Column 3	0.6	1.4	0.9
Column 4	0.5	1.5	2.0
Column 5	0.5	1.8	1.5
Column 6	0.4	1.0	1.3
Column 7	0.6	1.3	0.9
Ave	0.6	1.5	1.3
Std Dev	0.2	0.2	0.4
% Std Dev	34.9	16.8	30.2

- Months of behind-the-scenes work
- Values are not blank-subtracted
- Ratio between starting and electroformed copper expected to be much larger
- Led to analysis of electroforming bath solution using precipitation techniques
- Discussion on next slide!

Discussion of Results

- Calculation of method detection limit based on these data using standard t test for a 99% confidence interval (from 40 CFR 136)
  - Method detection limit = t for seven replicates * std. dev.
    = 3.18 * 0.2
    = 0.7 µBq 232Th /kg Cu
- Tracer (^{229}Th) yields with Cu about 2/3 blank levels - possible competition even with small Cu retention – doesn't limit measurement sensitivity
- Bath contamination found at relatively high levels (77 uBq/liter)
  - IGEX method used laborious multiple $CuSO_4$ recrystallizations
  - This batch only used one recrystallization
  - New method eliminates this source – creates $CuSO_4$ in situ
  - This result provides data point for Th rejection at lower concentrations – will be compared to upcoming tracer study
  - Implies about a factor of 100 Th rejection in process (at these Th levels)
- Statistically significant difference seen between blank and Cu samples
  - Lowest-uncertainty result is 0.9 ± 0.4 uBq/kg (starting anode)
  - Electroformed copper sample result is 0.7 ± 0.6 uBq/kg

Summary of ^{232}Th Assay

- Radiochemistry + ICP-MS for ^{232}Th in Cu
  - Method sensitivity
    - 1 σ = 0.2 uBq/kg
    - 99% CL = 0.7 uBq/kg
  - Unexpected bath contamination at 77 uBq/liter found, mitigated by new bath setup procedure
  - Lowest positive measurements in UHP copper
    - 0.9 ± 0.4 uBq/kg (starting anode)
    - 0.7 ± 0.6 uBq/kg (electroformed copper)
    - Publication in preparation
  - Two more campaigns of seven-fold-replicate method planned in FY07, each 2-3 samples

^{228}Th Tracer Study

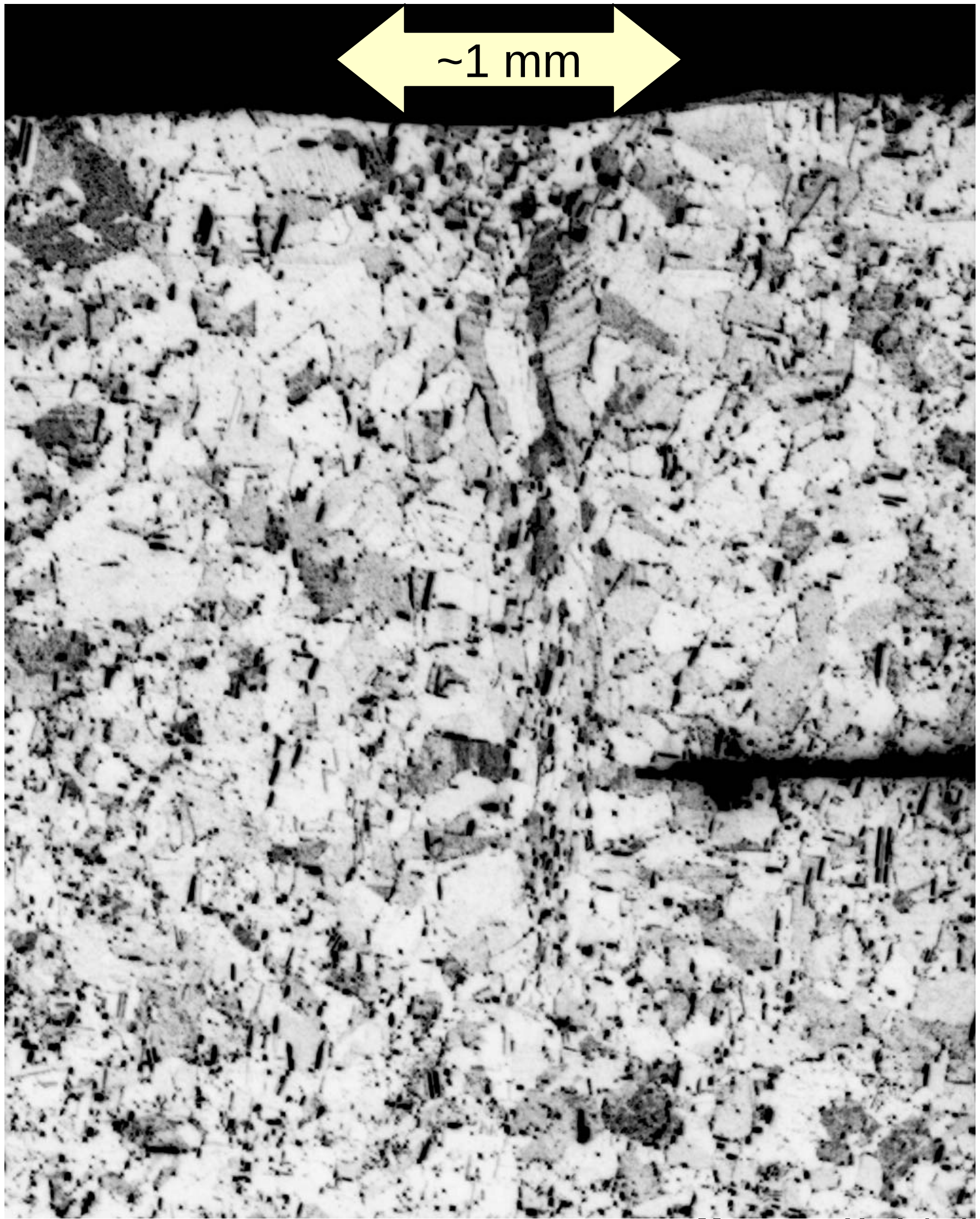
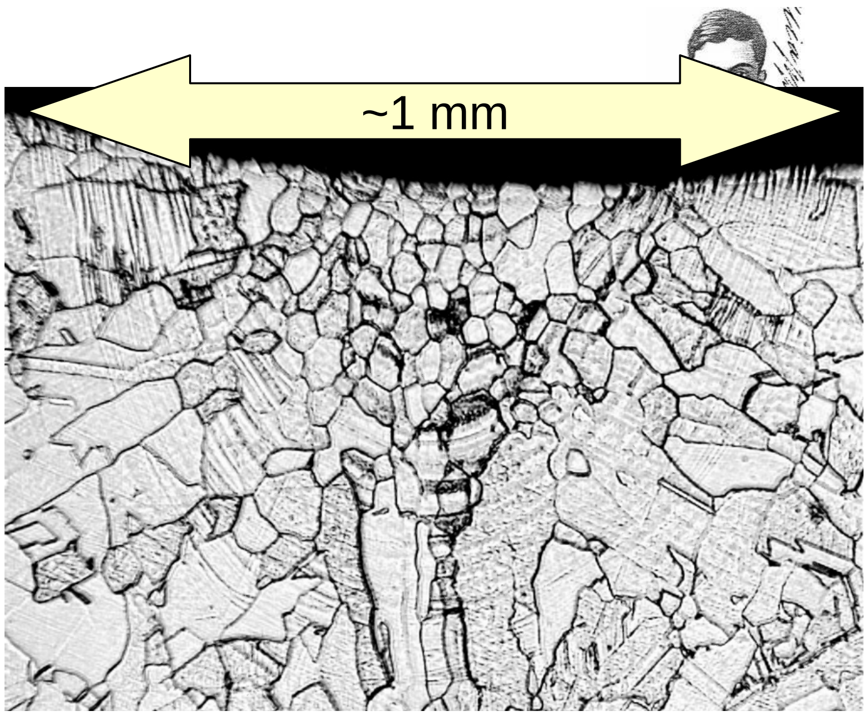
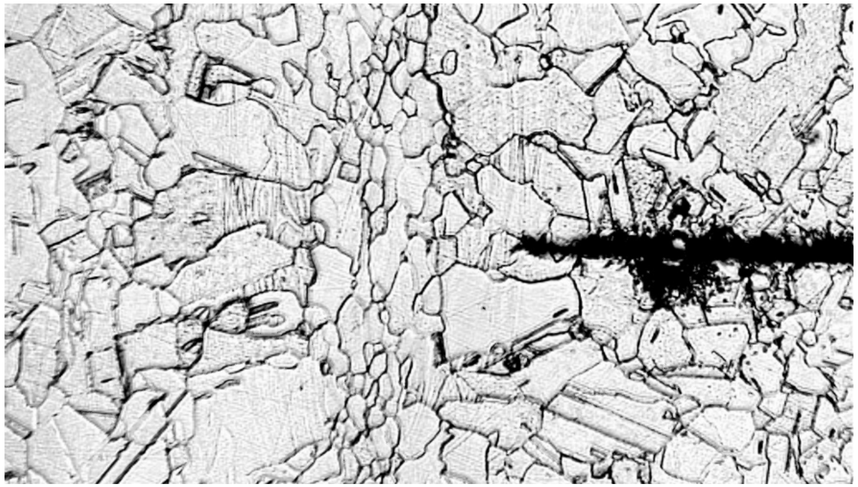
- Goal is to study behavior of Th at atom concentrations near Majorana target purity
- High specific activity means above-ground low-background HPGe can measure target tracer concentrations in ~1-gram electroformed copper samples
  - Can radiometrically measure atom concentration equivalent to <0.5 uBq/kg ^{232}Th
  - Expect to cover 5 or 6 orders of magnitude
- Status
  - 5 mCi ^{228}Th source received from ORNL
  - Experimental plan developed
    - Includes plans to look at broken equilibrium, etc.
  - "Cold" experiments happening now
    - Students + HRA + dispersible source = proceed carefully

Electron-Beam Welding

Majorana Update to GERDA
June 10 - 14, 2007, IRMM, Belgium

Electron-Beam Welding Tests

- **E-Beam Welding**
  - **Clean process – done in vacuum**
  - **No additional materials**
  - **Very good process control**
  - **Used on GeMPI detectors**
  - **Affordable cost**
  - **Potentially much faster than IGEX electro-welding method**
- **Current tests performed**
  - **First work with PNNL E-Beam group**
  - **Established welding parameters**
  - **Use simple tube geometry to facilitate vacuum tests of wield**
  - **Sectioning and microscopy for weld penetration, crystallography studies**

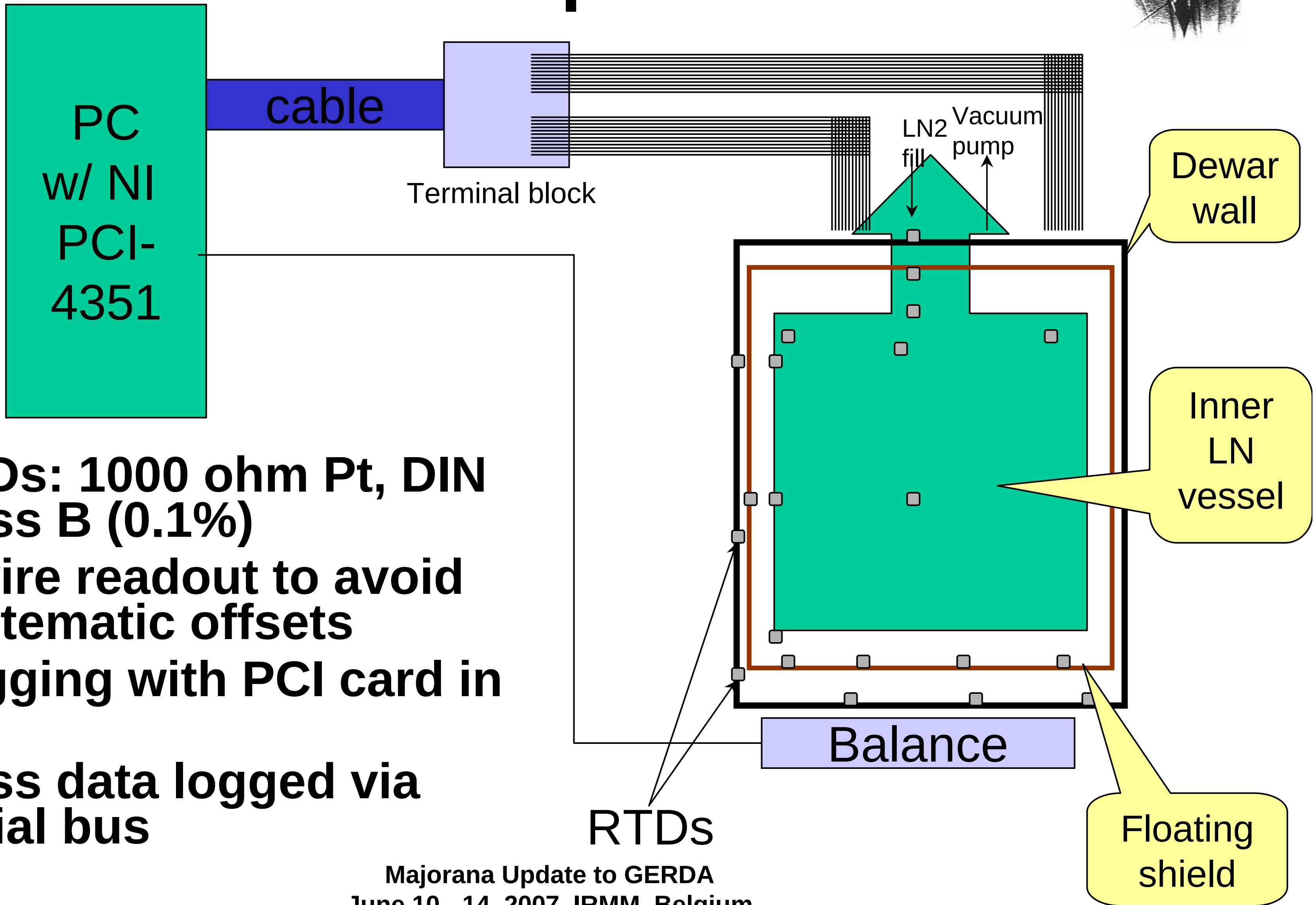

~1 mm

~1 mm


Copper Emissivity Measurements

Copper Emissivity Measurements

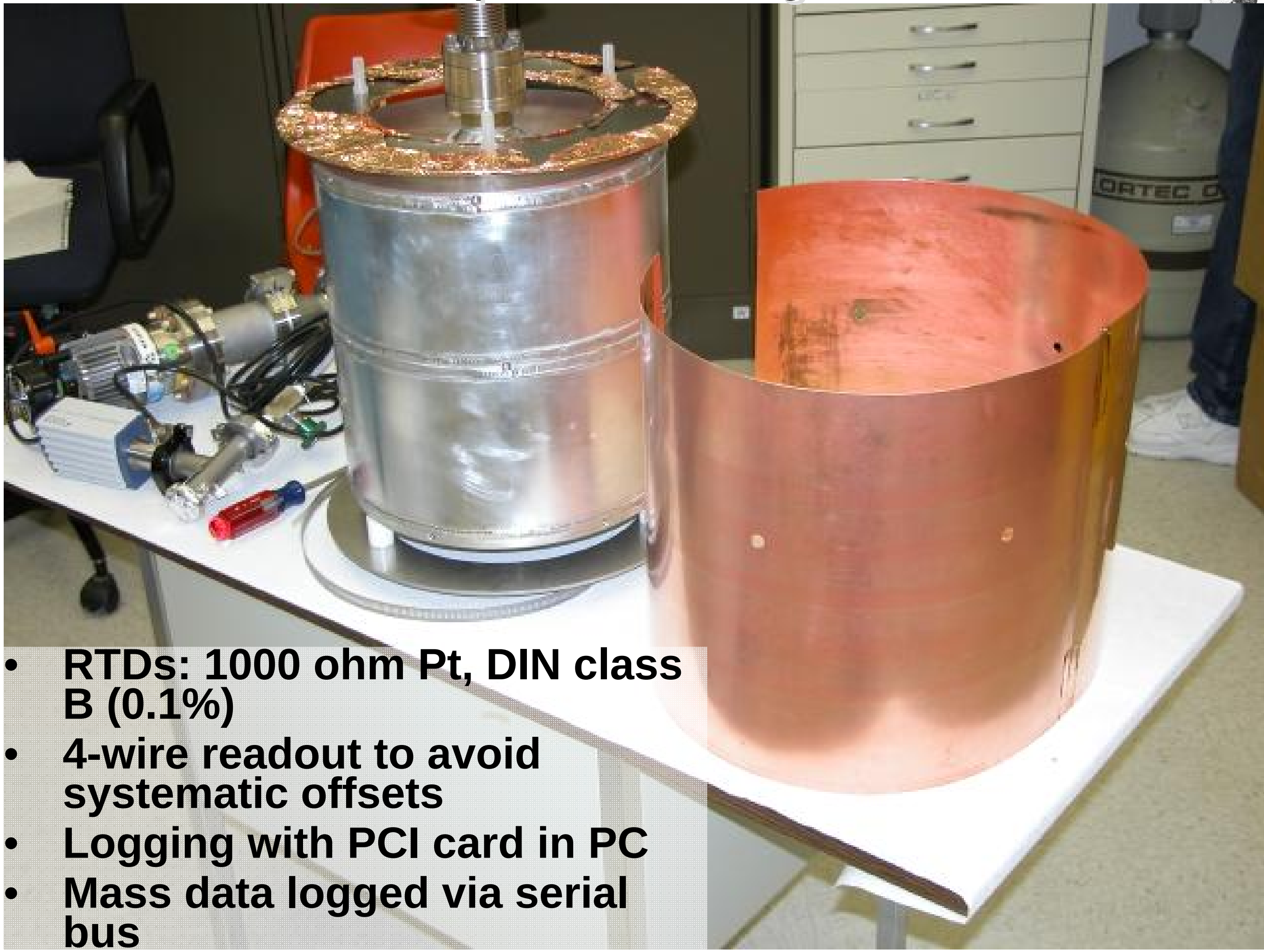
- **Goals**
  - **Control emissivity of clean copper surfaces**
  - **Demonstrate practicality of using floating copper shield to reduce heat loads**
- **Method**
  - **Borrow “cryogenic test setup” from another project**
  - **Clean and passivate copper lining and floating shield, measure cryogenic power load**

Schematic of cryogenic test setup



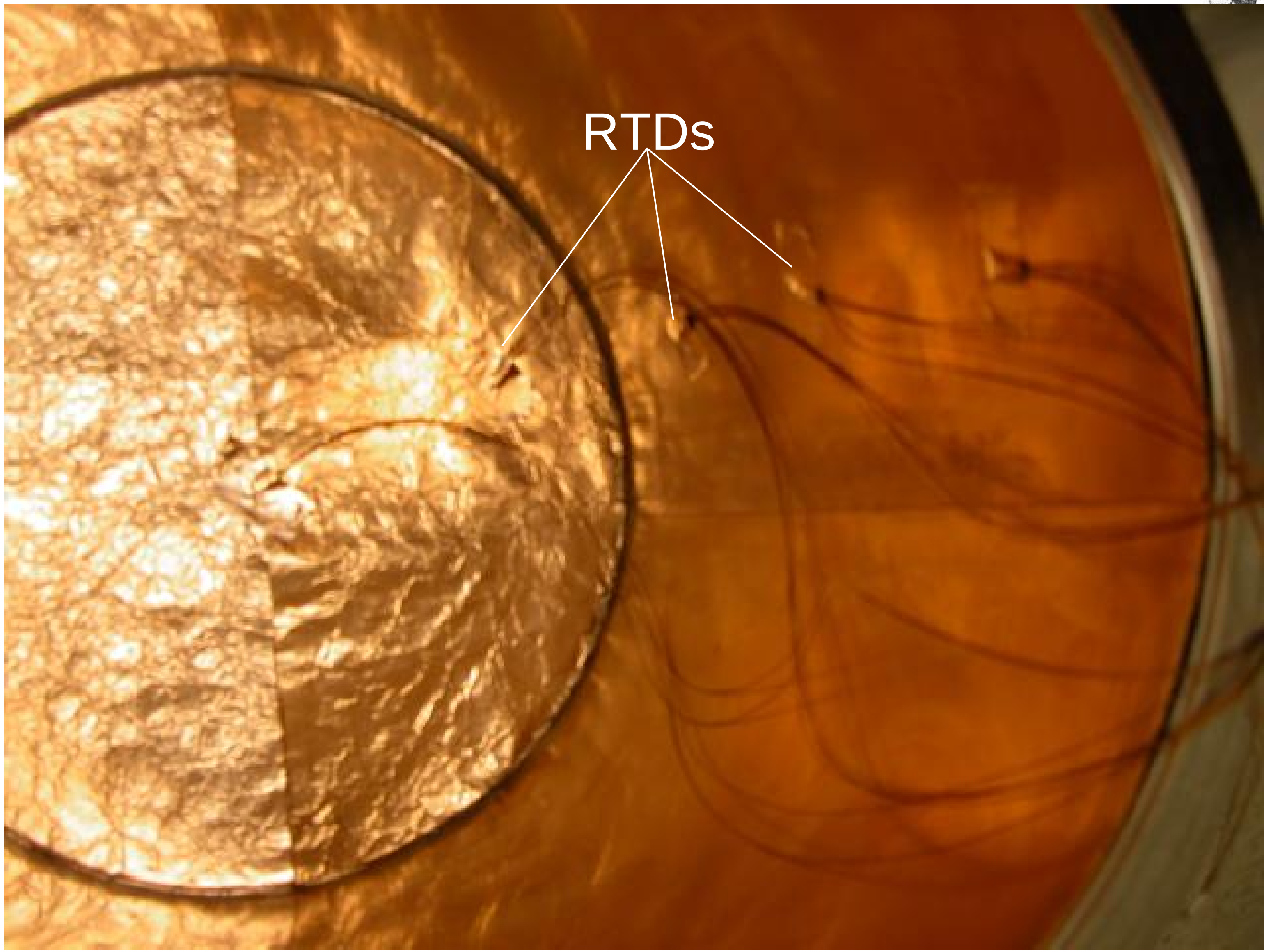
- **RTDs: 1000 ohm Pt, DIN class B (0.1%)**
- **4-wire readout to avoid systematic offsets**
- **Logging with PCI card in PC**
- **Mass data logged via serial bus**

Dewar and part of floating IR shield

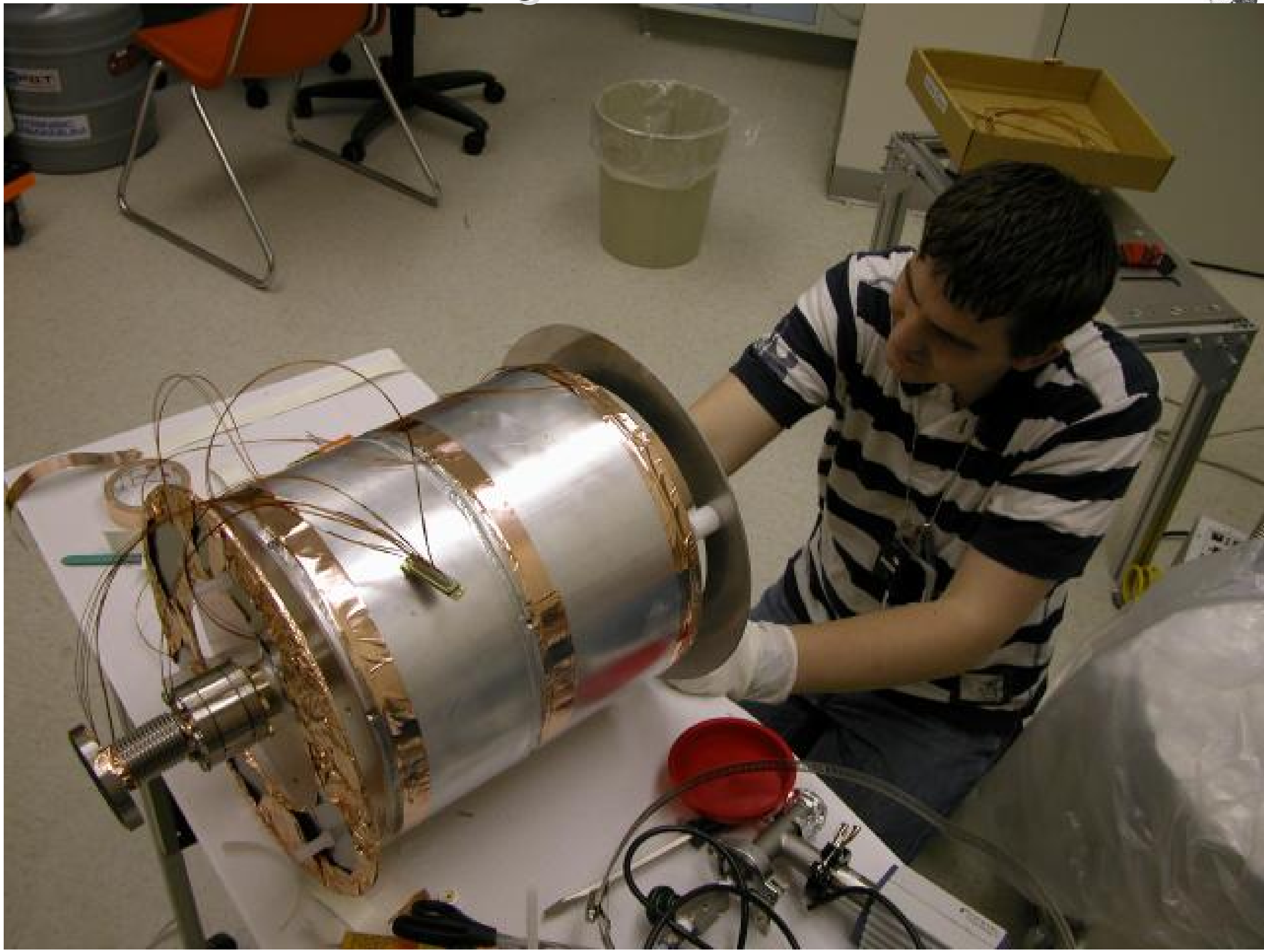


- **RTDs: 1000 ohm Pt, DIN class B (0.1%)**
- **4-wire readout to avoid systematic offsets**
- **Logging with PCI card in PC**
- **Mass data logged via serial bus**

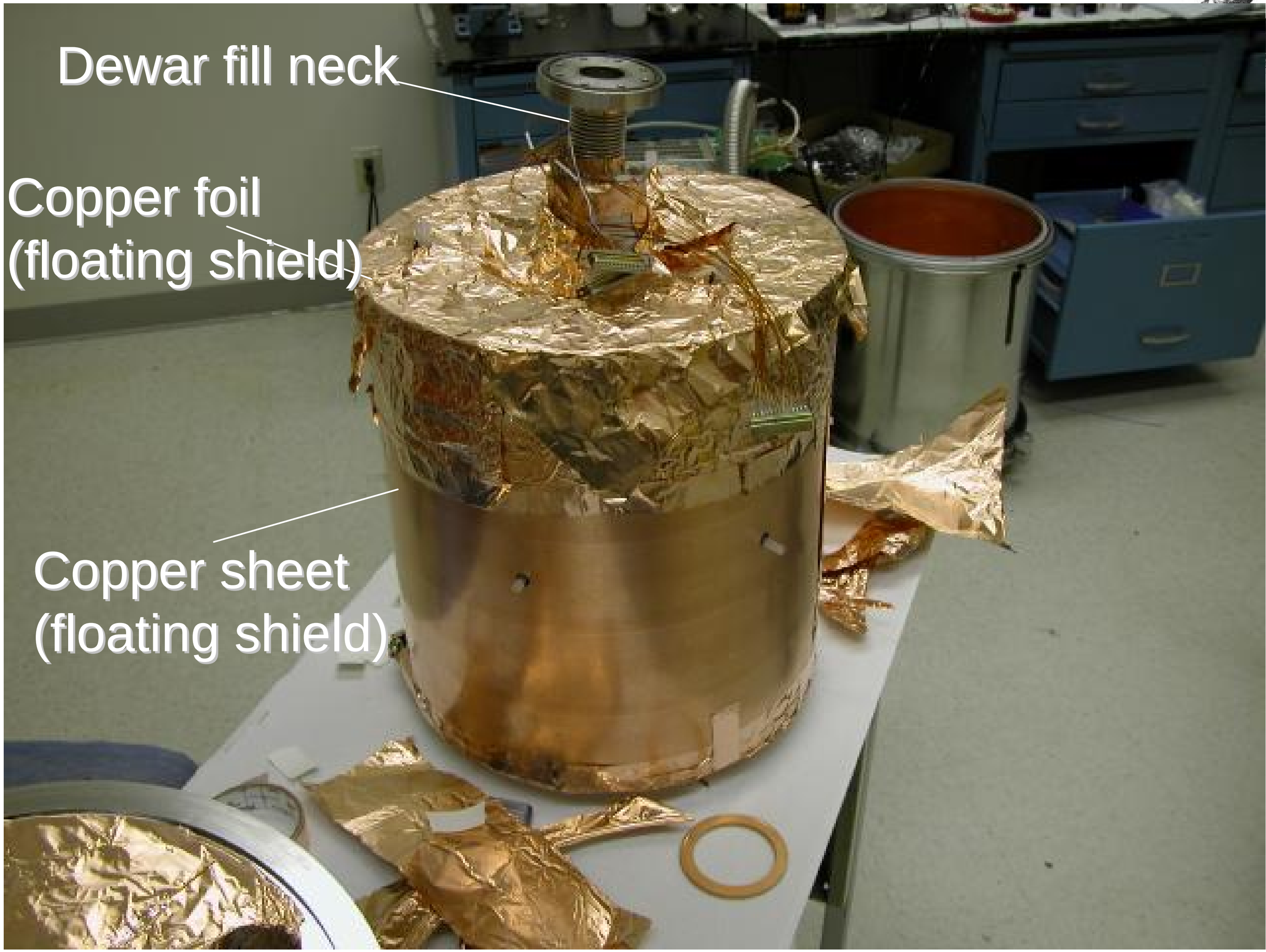
Inside of vacuum chamber



Mounting RTDs to dewar



Assembled floating IR shield



Lining lid of chamber

Copper Surface Preparation

- **Goal: Low emissivity**
- **Highly polished copper : ε~2-3%**
- **Oxidized copper : ε~10-80%**
- **Most copper cleaned and passivated with previously established method, Hoppe *et al.****
    - **Acidified peroxide etch (3% H_2O_2, 1% H_2SO_4)**
    - **Citric acid passivation (~1%)**
- **Small amount of copper foil only passivated with more concentrated (5%) citric acid due to time constraints**

*Nucl. Instr. Meth. **A**, in press (2007)

Cryogenic Test System: Measured temperatures

Outer wall:
T=293K

Floating IR shield:
T=233K

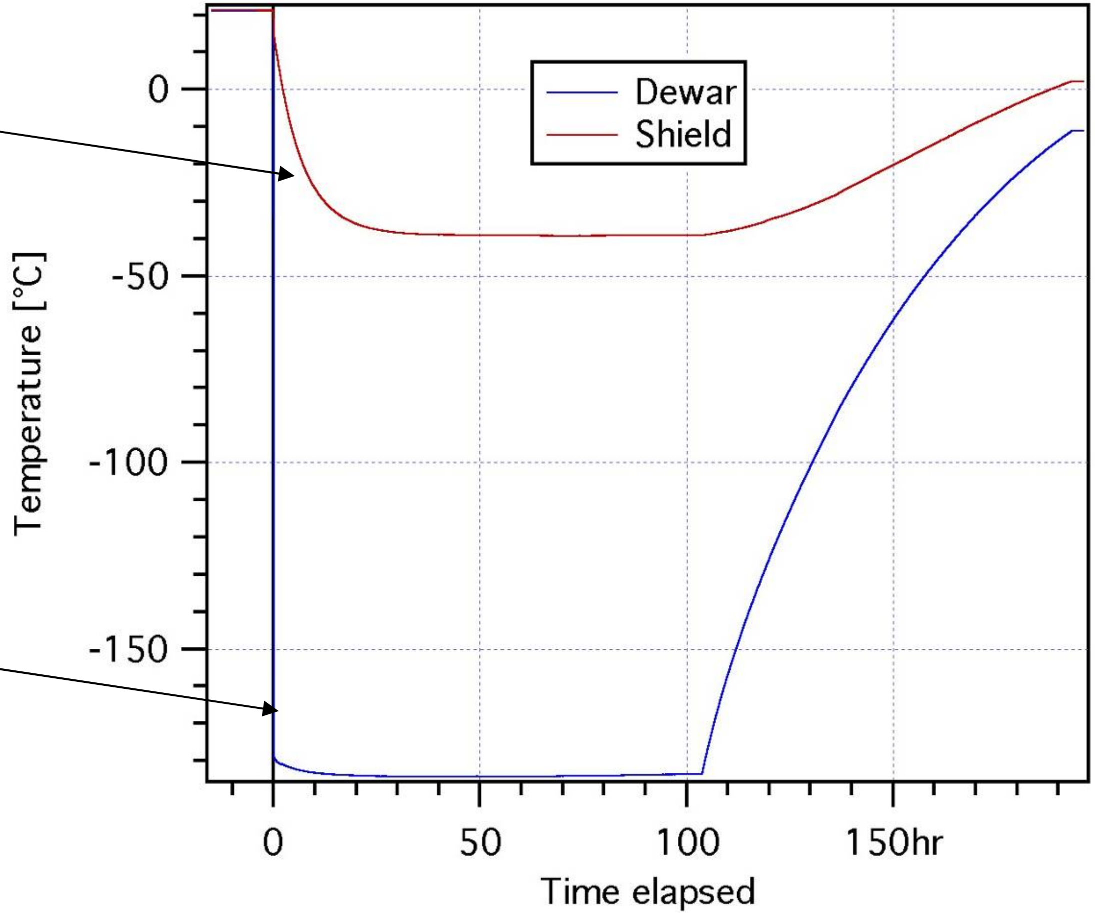
Dewar:
T=83K

Temperature measurements

8 liters of LN

24-hr cool-down time for floating shield

Immediate cool-down for inner vessel

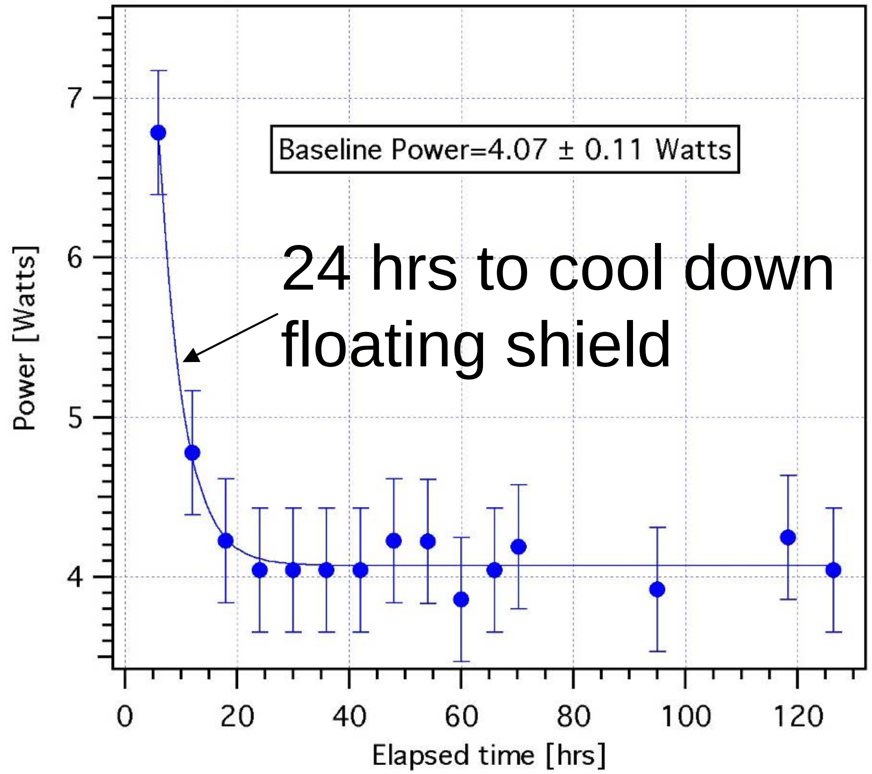


(data taken with incomplete IR shield - no coverage at bottom)

Cooling Power

Final configuration of cryogenic test assembly

- **Power calculated from LN boiloff rate**
- **Gradual power reduction understood: result of slow heat extraction from materials (SS, Cu) in the floating shield**
- **Required about half the cooling power of non-shield design**



Emissivity Measurements

- **Use LN boiloff rate, material properties, and the following equations to extract emissivity:**

$$P_{total} = P_{rad} + P_{con}; \quad P_{con} = \frac{kA\Delta T}{d}; \quad P_{rad} = \frac{\sigma\left(T_1^4 - T_2^4\right)A_1}{\frac{1}{\varepsilon_1} + \left(\frac{1}{\varepsilon_2} - 1\right)\frac{A_1}{A_2}}$$

- **Conductive heat load = ~1.4 Watts, radiative heat load ~2.7 Watts**
- **Hoppe passivated copper: ε=2.5(2)%**
  - **Consistent with earlier, small scale measurements**
  - **Passivation technique produces consistent results**
  - **As good as highly-polished copper and almost as good as gold**
- **“Other” passivated copper: ε=3.7(4)%**

Copper Emissivity Summary

- Goals
  - Control emissivity of clean copper surfaces
  - Demonstrate practicality of using floating copper shield to reduce heat loads
- Emissivity
  - Demonstrated emissivity of 2.5% is excellent result even when compared to gold
  - Small scale and large scale measurements agree
- Floating shield
  - Reduced heat load in cryogenic test assembly by about a factor of two, consistent with modeling

Summary

- Copper Electroforming Status
  - PNNL above-ground labs
    - Increased capacity
  - LANL underground work at WIPP
    - First test piece electroformed underground
- Electroformed Copper Purity
  - ^{232}Th Assay
    - New method sensitivity 0.7 uBq/kg (90% CL)
  - ^{228}Th Tracer Study
- Electron-Beam Welding
- Copper Emissivity Measurements
  - Passivation providing stable surface at 2.5% emissivity – "good as gold"