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Radioactive Background Control at Pacific Northwest National Laboratory

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20th October 2026

CPAD 2026

University of Washington, Seattle, USA

U.S. DEPARTMENT of ENERGY BATTELLE

PNNL is operated by Battelle for the U.S. Department of Energy



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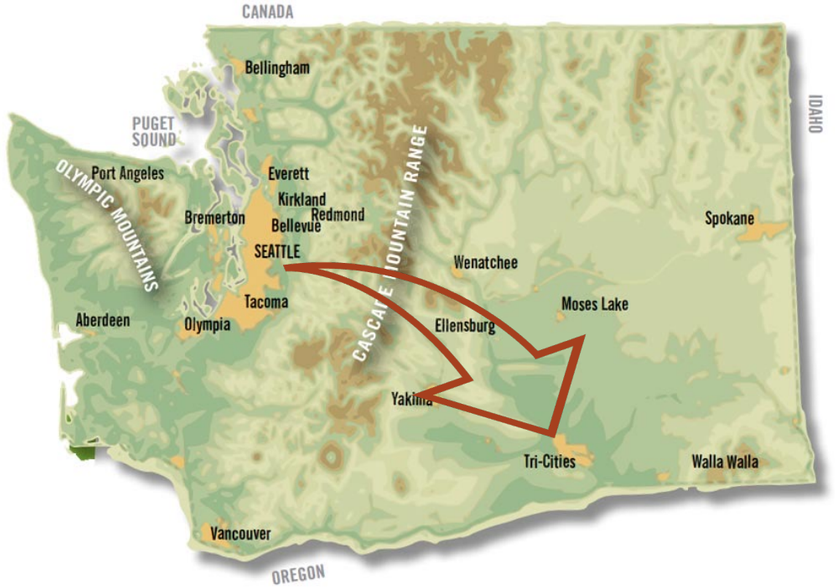
U.S. DEPARTMENT of ENERGY
National Laboratory

Located in southeast Washington state
(~ 3.5 hrs from Seattle)

Founded in 1965
Origins trace back to Hanford site for plutonium production in World War II

~ 6000 staff members

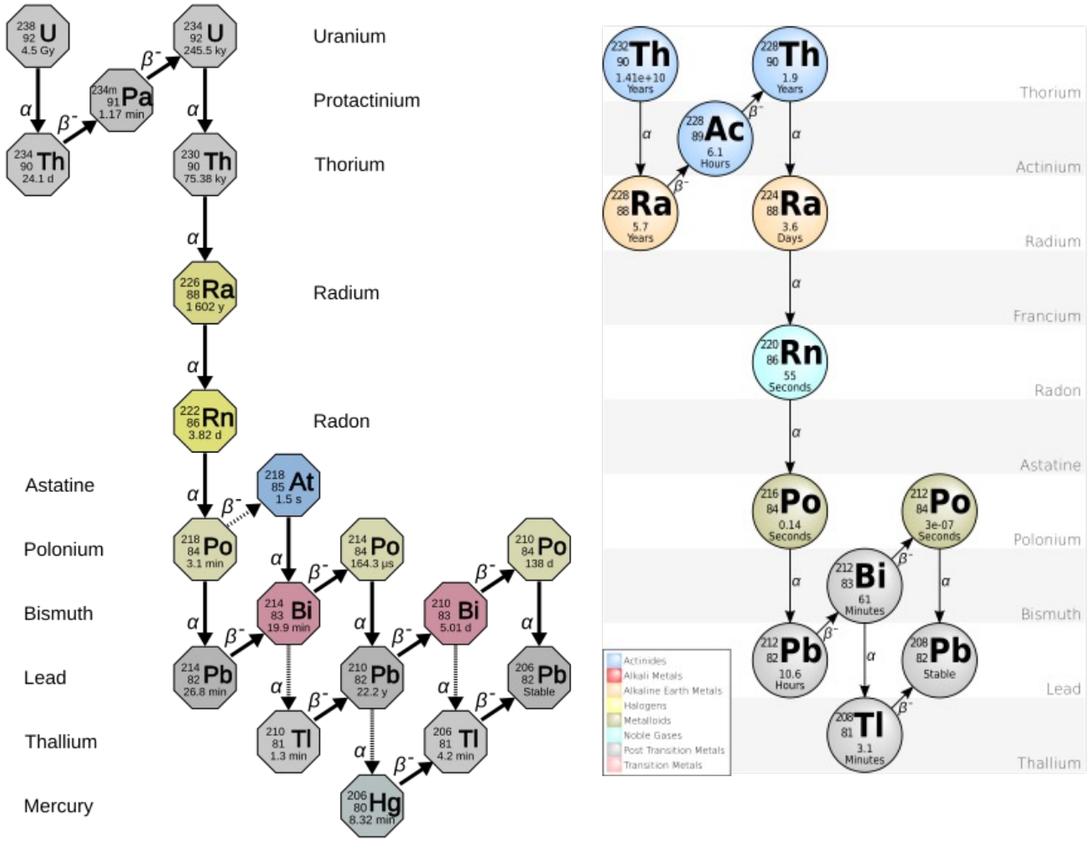
PNNL's core capabilities :
Chemical and Material Sciences
Computational and Mathematical Sciences
Earth and Biological Sciences



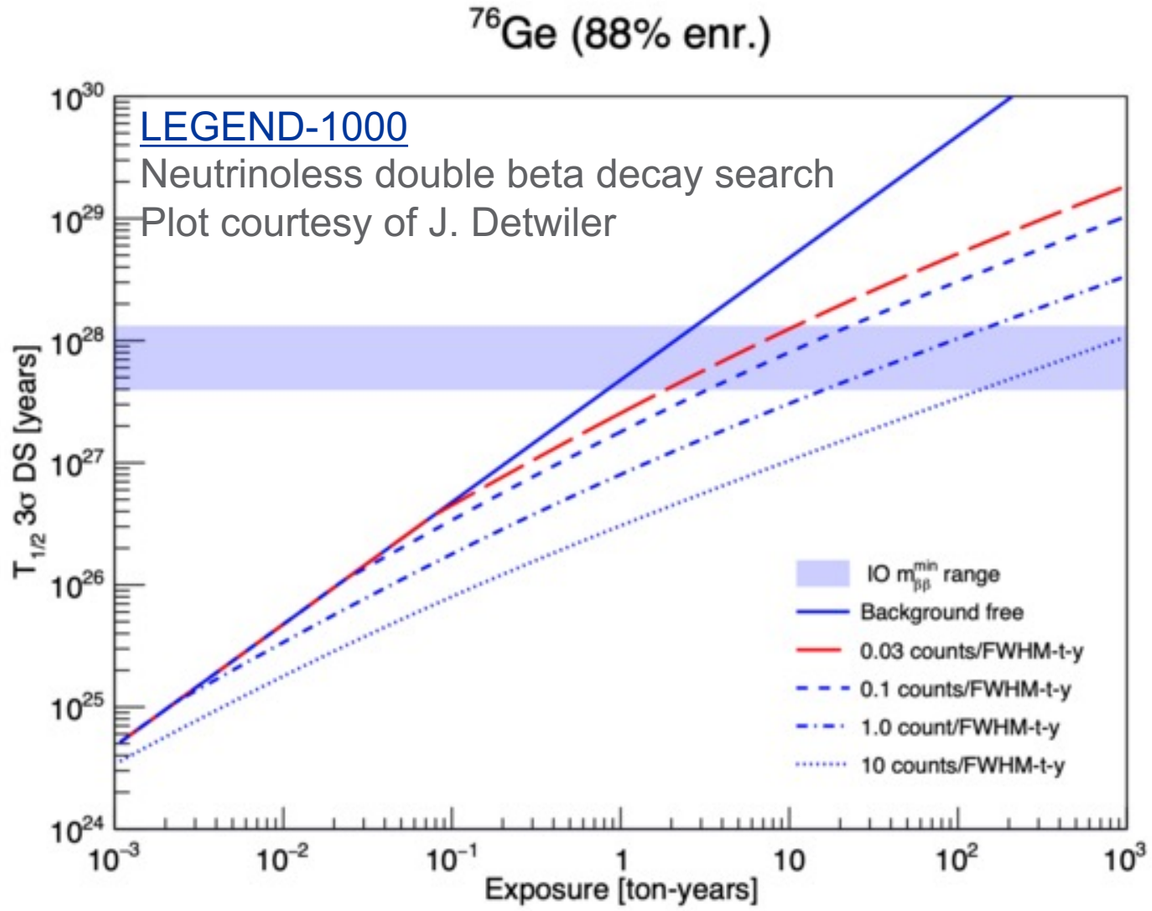


Radioactive Backgrounds

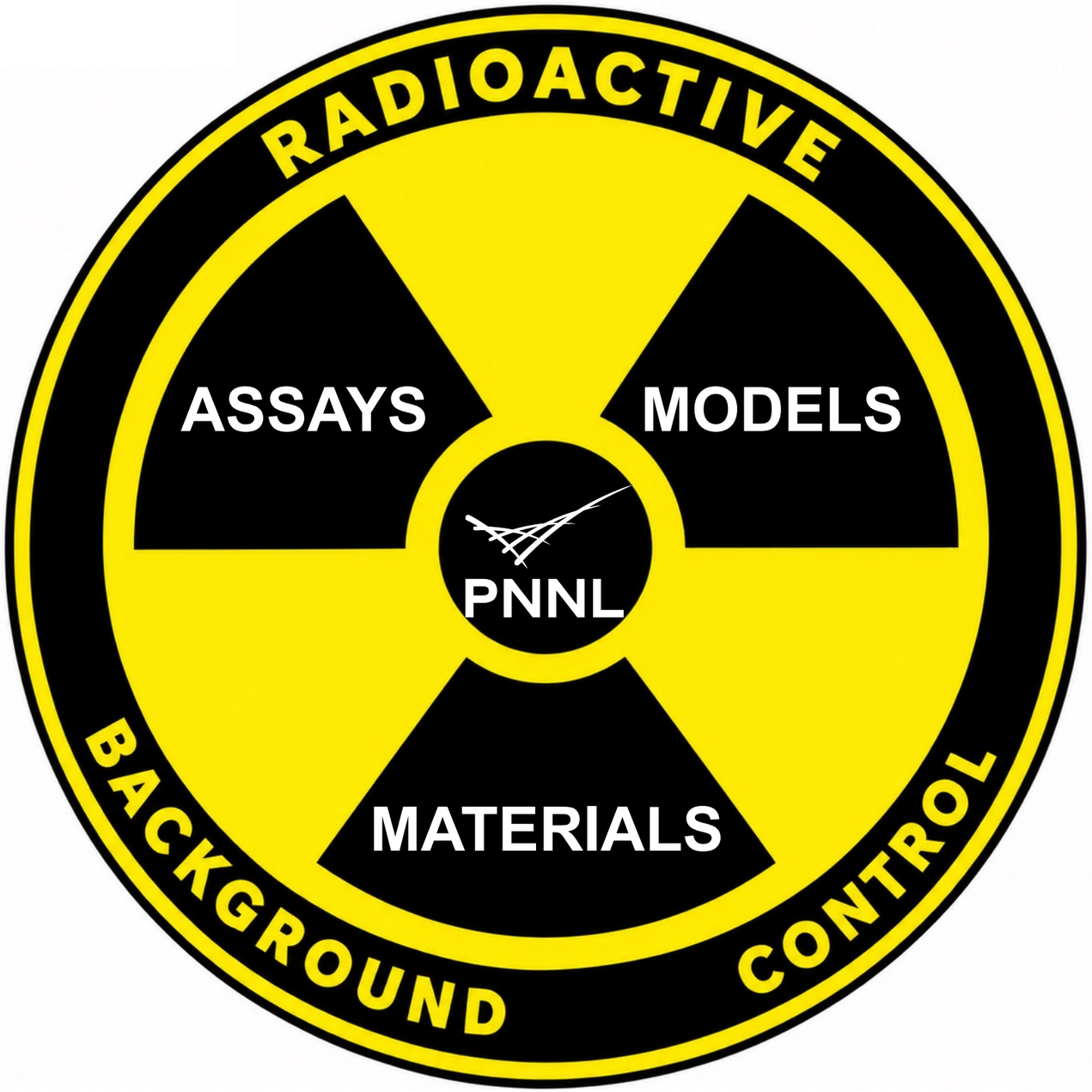
Radioactive backgrounds are **the biggest challenge** to rare event searches in the keV-MeV range



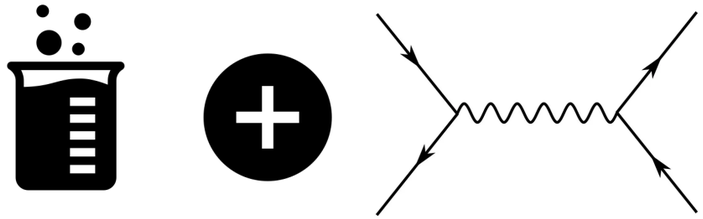
Primordial ^{238}U and ^{232}Th decay chains



Measure, Model, Mitigate



Strong collaboration between chemists and physicists

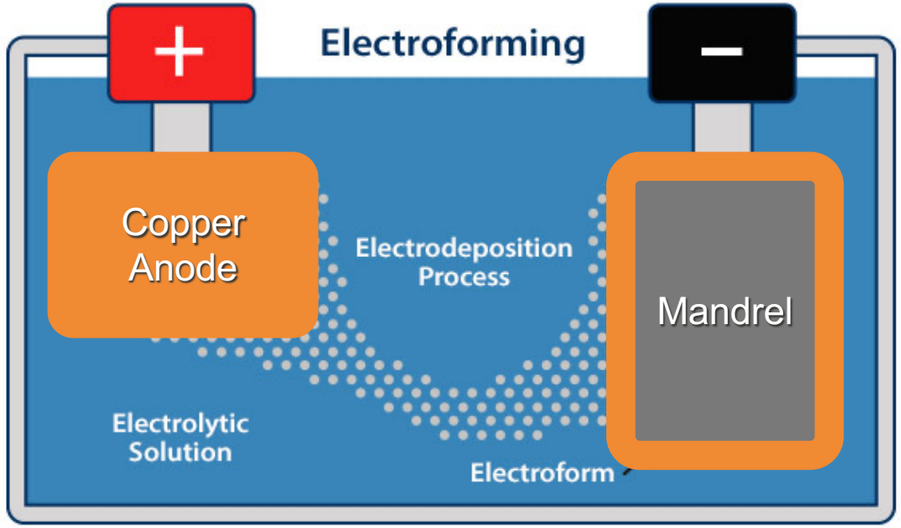


“Having a physics PhD doesn’t mean you know much physics, but it often means you don’t know any chemistry”

ULB Electroformed Copper

Reductants		Oxidants	E^0 (V)
$Cu^{2+} + 2e^-$	$\leftrightharpoons$	Cu	+0.34
$Pb^{2+} + 2e^-$	$\leftrightharpoons$	Pb	−0.13
$U^{3+} + 3e^-$	$\leftrightharpoons$	U	−1.80
$Th^{4+} + 4e^-$	$\leftrightharpoons$	Th	−1.90
$K^+ + e^-$	$\leftrightharpoons$	K	−2.93

- Electroforming takes advantage of the **relatively high reduction potential of copper compared to most radioisotopes**
- By carefully setting the electroplating voltages and bath parameters, pure copper can be plated from solution, leaving radiocontaminants in the electrolyte.
- PNNL chemists have developed custom electroformed copper that has been demonstrated to have **< 10 parts-per-quadrillion U & Th contamination, > 25x lower than the cleanest commercial copper**



	^{238}U [ppt]	^{232}Th [ppt]
Commercial Copper	0.25	0.13
Custom Electroformed Copper	< 0.01	<0.01

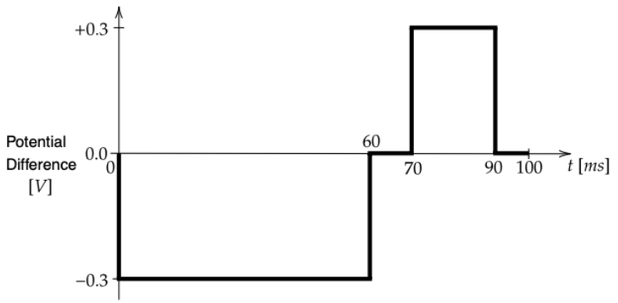
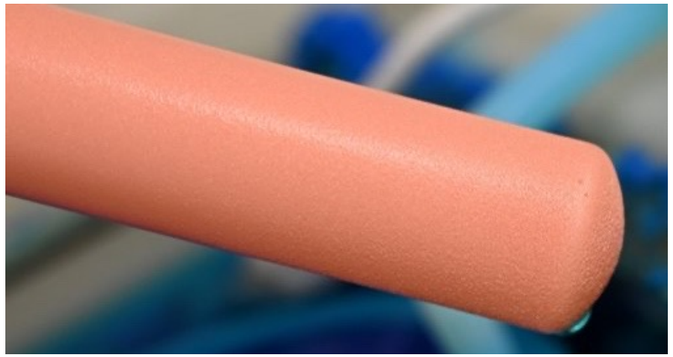


Figure 5: Waveform used in the electroplating. The negative terminal was attached to the cathode.



Electroformed Copper

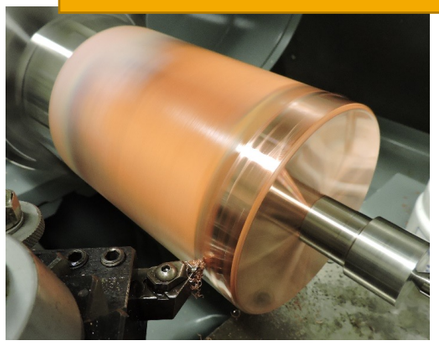
- Custom electroformed copper in various shapes and sizes has now been deployed in multiple rare event experiments including Majorana Demonstrator, SuperCDMS, NEWS-G, DAMIC-M, etc.
- On-going R&D includes large **scalable bath designs, alloys for improved strength, and automated bath control to improve copper properties and reproducibility**







IR Shields & Vacuum Jackets





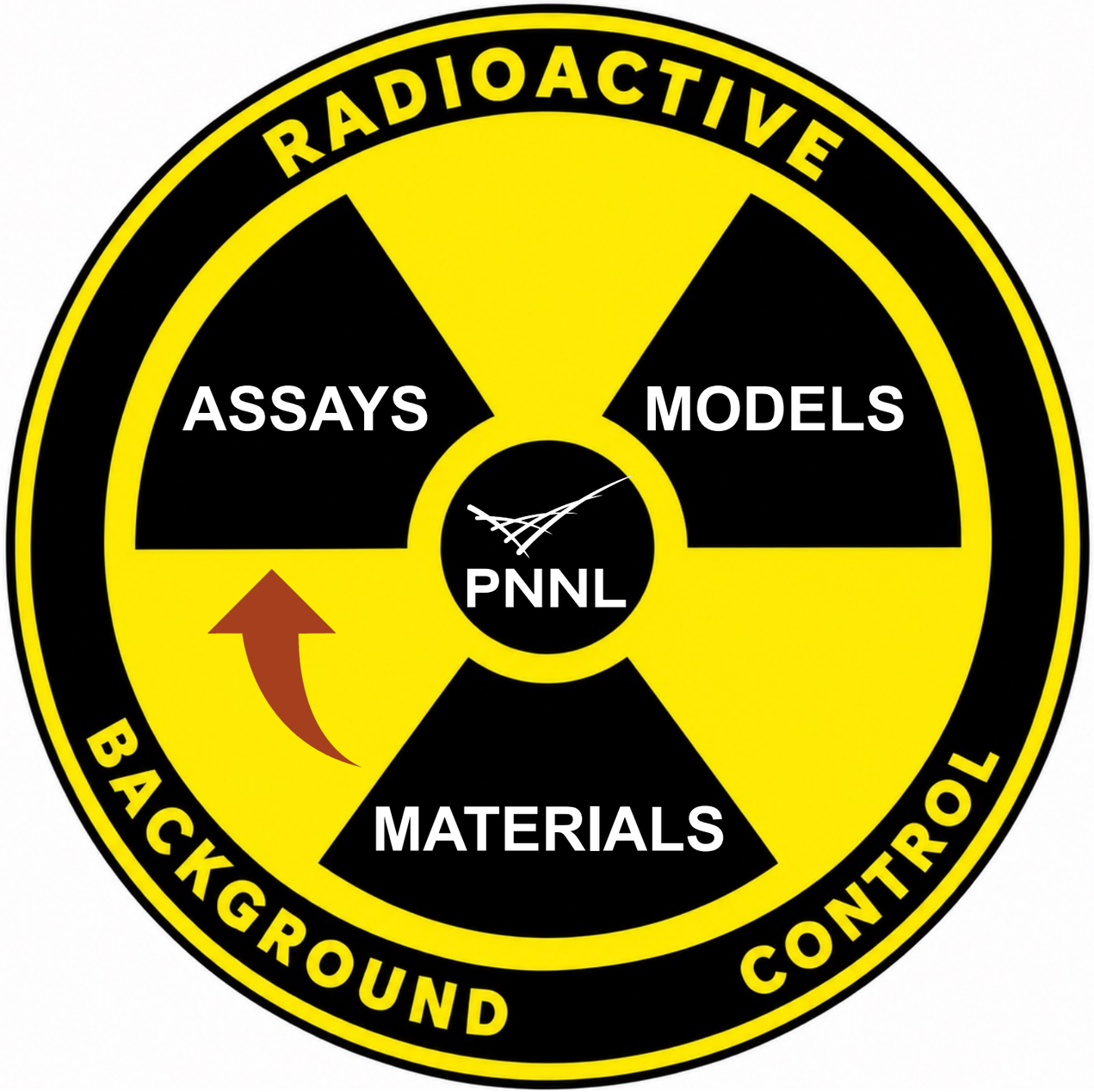
1” diameter gas proportional counters



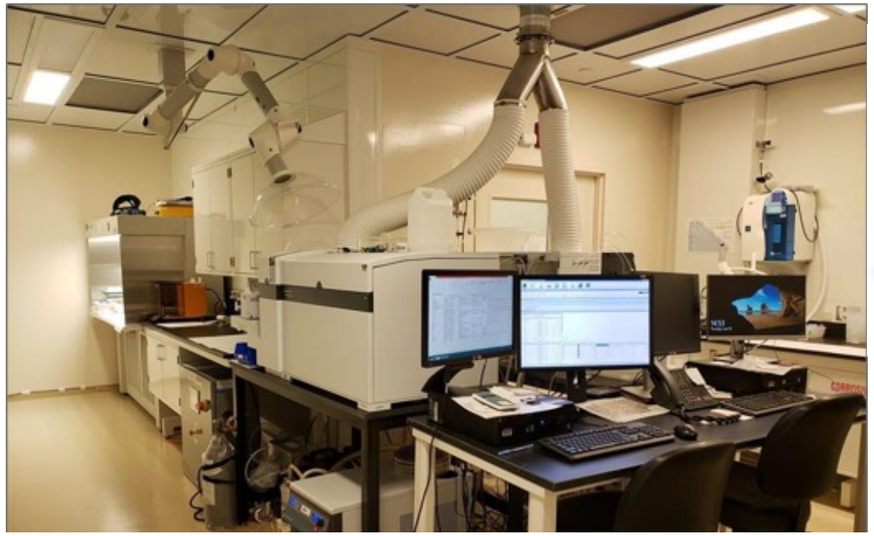


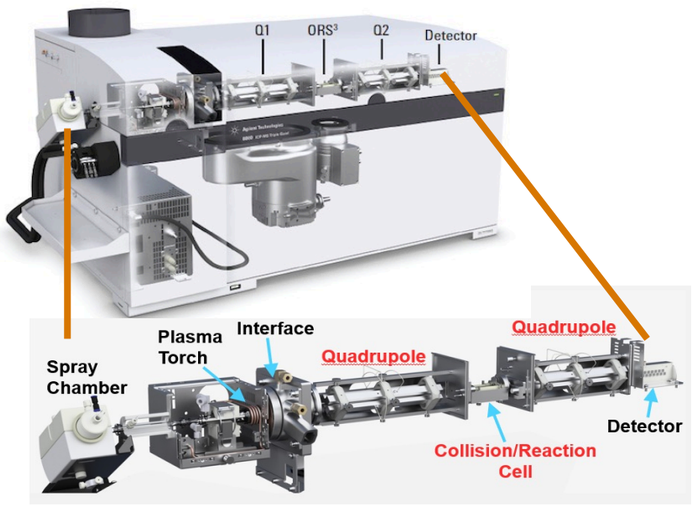
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Custom electroformed copper has such low levels of radiocontamination that it **triggered the development of ultra-sensitive (sub-parts-per-trillion) assay techniques at PNNL**



Facilities:
Ultra-sensitive material assay laboratory

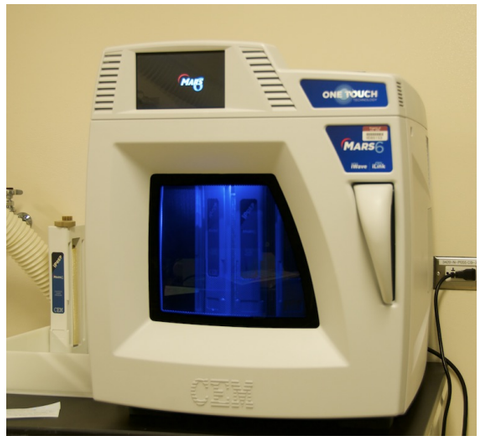


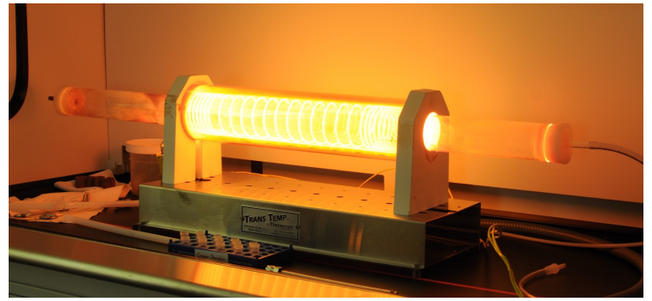


- Class 10,000 (measured at ≈1000) clean chemistry laboratory
- **Two dedicated triple-quadrupole ICP-MS instruments** (Agilent 8800 and Agilent 8900)
- Microwave and dry ash sample digestion capabilities
- Milli-Q water system
- Optima grade regents
- Low-background PFA labware









Ultra-sensitive material assay methodologies

To achieve sub-ppt sensitivity, the process is more important than the instrument

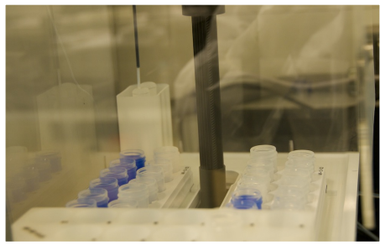
- Meticulous labware cleaning and validation protocols
- Samples prepared and assayed in triplicate
- Process blanks (n≥3) used to establish detection limits *each time*
- Quantitation of ^{232}Th and ^{238}U sensitivity using non-natural ^{229}Th and ^{233}U tracers

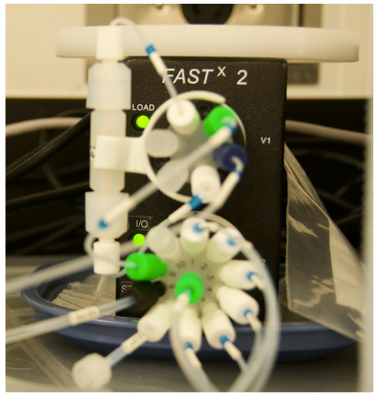
For 10's of parts-per-quadrillion sensitivity:

- Chemical separation of matrix using anion exchange
- Pre-concentration of analytes
- **Automated** separation system to improve reproducibility and throughput



2DX prep*FAST* Station





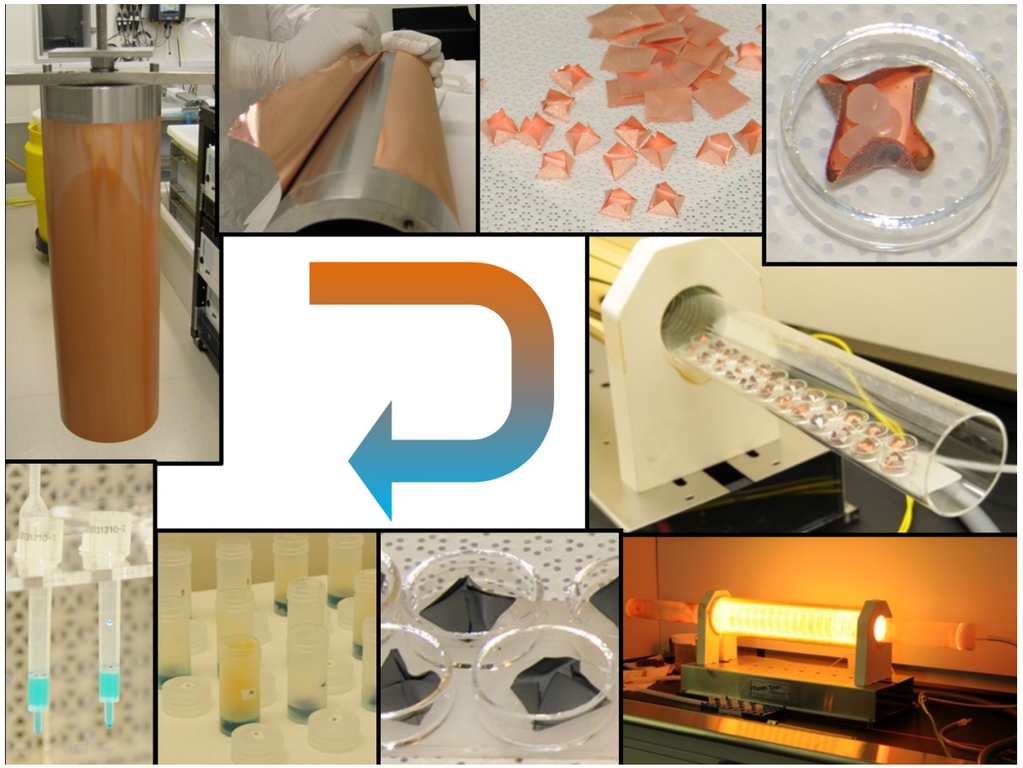
- ^{232}Th < 3.6 fg/g (< 0.015 μBq/kg)
- ^{238}U < 9.4 fg/g (< 0.12 μBq/kg)

I.J. Arnquist, M.L. di Vacri, E.W. Hoppe, NIM A 965 (2020) 163761

Advancing assay methodologies

Clean dry ash methodology for ultra-sensitive assay of **polymeric materials**

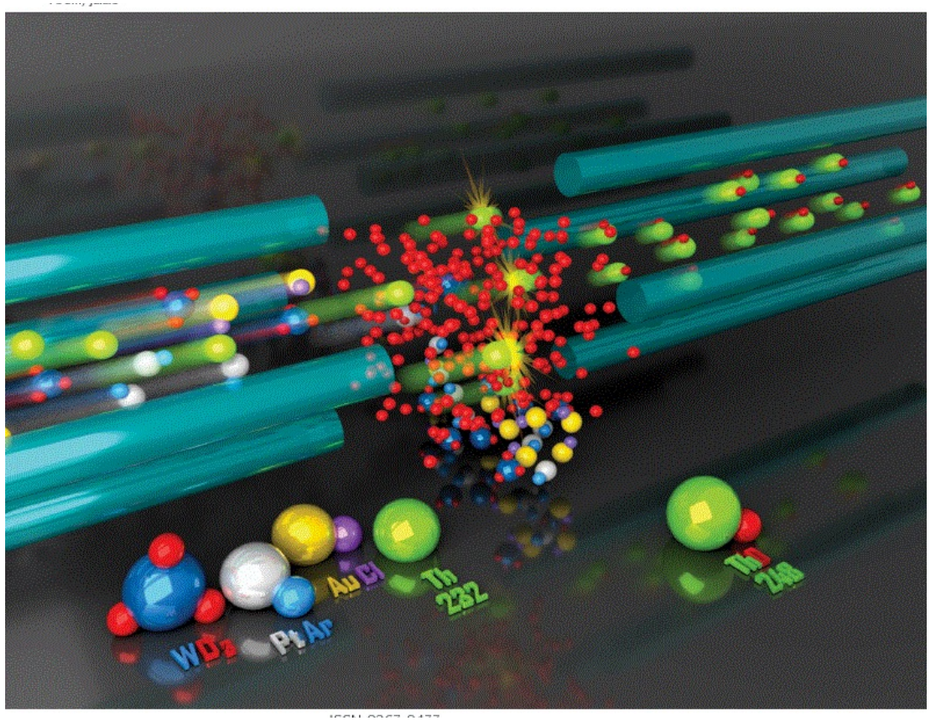
- Uses EFCu as a clean “container”



I.J. Arnquist, E.W. Hoppe, Bliss M., Grate JW., Anal. Chem. (2017) 89 (5):3101-3107

Reducing interferences

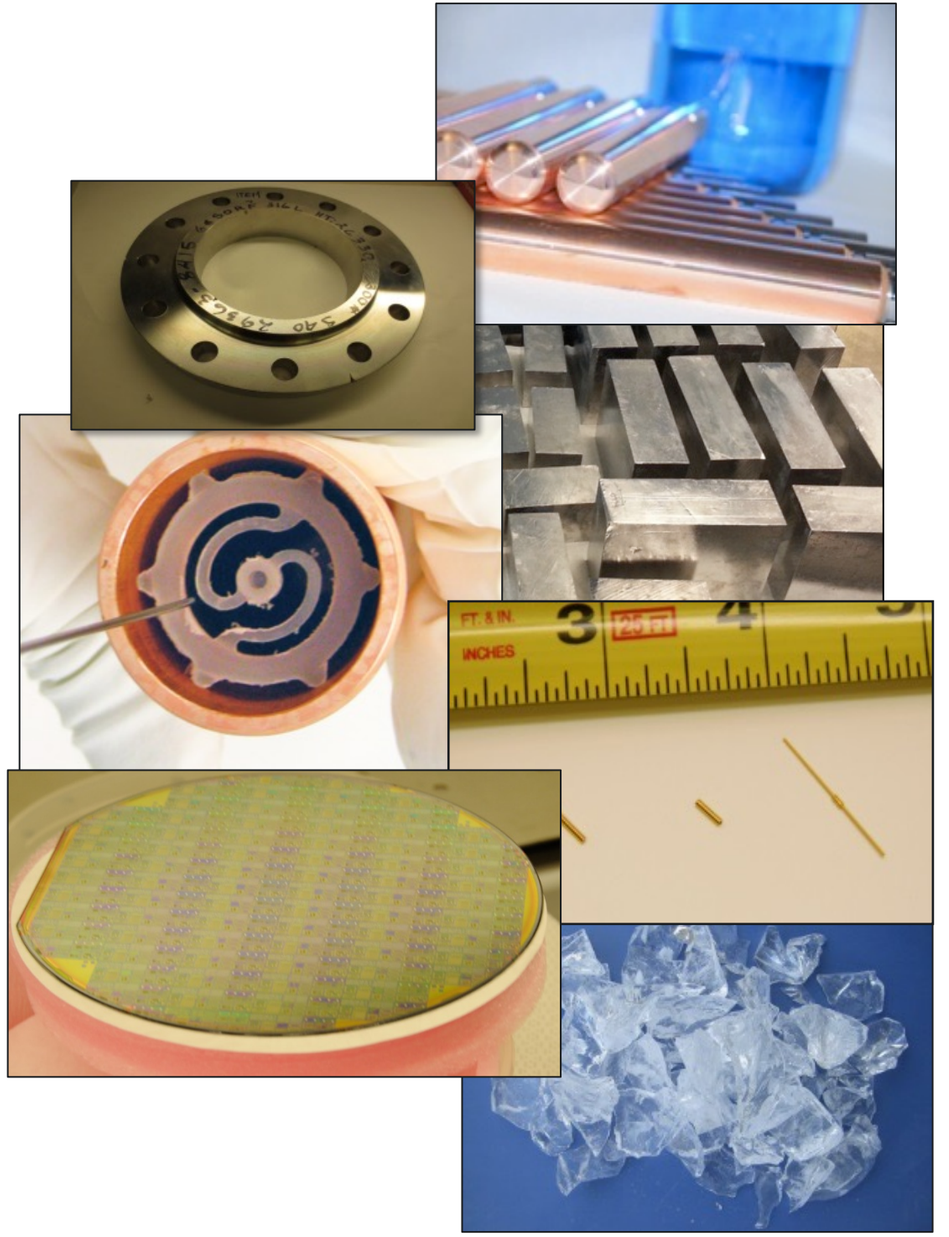
- Polyatomic interferences can limit ICP-MS sensitivity (e.g. AuCl, WO_3, Mo_2Ar)
- Using a reaction gas (e.g. O_2) can mass shift the isotopes of interest to an interference-free m/q region



K. Harouaka, E. Hoppe, I. Arnquist, J. Anal. At. Spectrom. (2020) 35 (12): 2859–2866

PNNL ICP-MS Sensitivities

Material	Th/U Detection Limits	
	µBq/kg	ppt
Copper†	<0.1	<0.01
Titanium, Steel, Brass, Bronze, Lead	<1	<0.1
Polymers‡ (HDPE, PTFE, Acrylic, Kapton®, *etc.*)	<1	<0.1
Linear Alkyl Benzene (LAB)	<0.1	<0.01
Silicon, Quartz, Fused Silica	<1	<0.1
Cables and Connectors	<1	<0.1
Electronic Components (resistors, capacitors, *etc.*)	<0.1 pg/piece <1 nanoBq/piece	
Solutions	<0.01	<0.001



1 ppt Th = 4.1 µBq ^{232}Th/kg
1 ppt U = 12.4 µBq ^{238}U/kg

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Measuring the radioactive contamination in detector components is just one aspect of background control

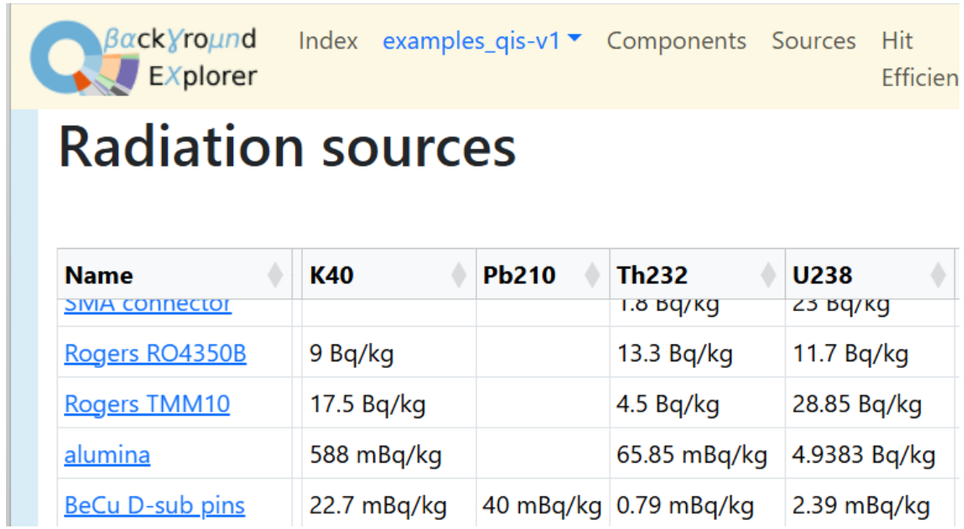
A **comprehensive background model** combines assay results, detector geometry, and particle tracking simulations to produce a detailed picture of the backgrounds that can be used to fit data, evaluate sensitivity and highlight critical areas for improvement.

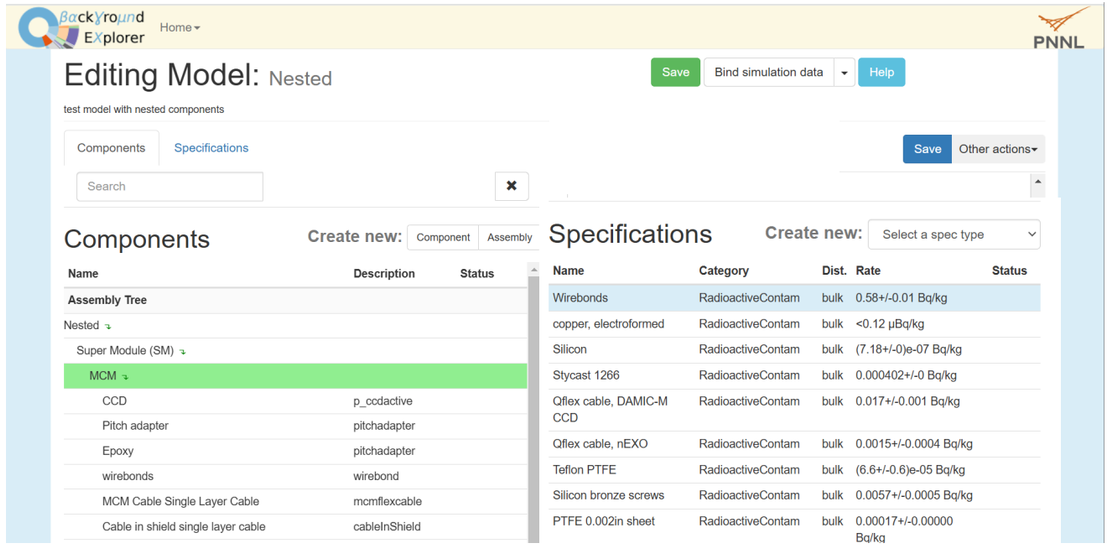


Background Modeling

Background Explorer:
Open-source, experiment-agnostic, software package for **combining assay results, particle tracking simulations and detector geometry to produce a comprehensive background model**

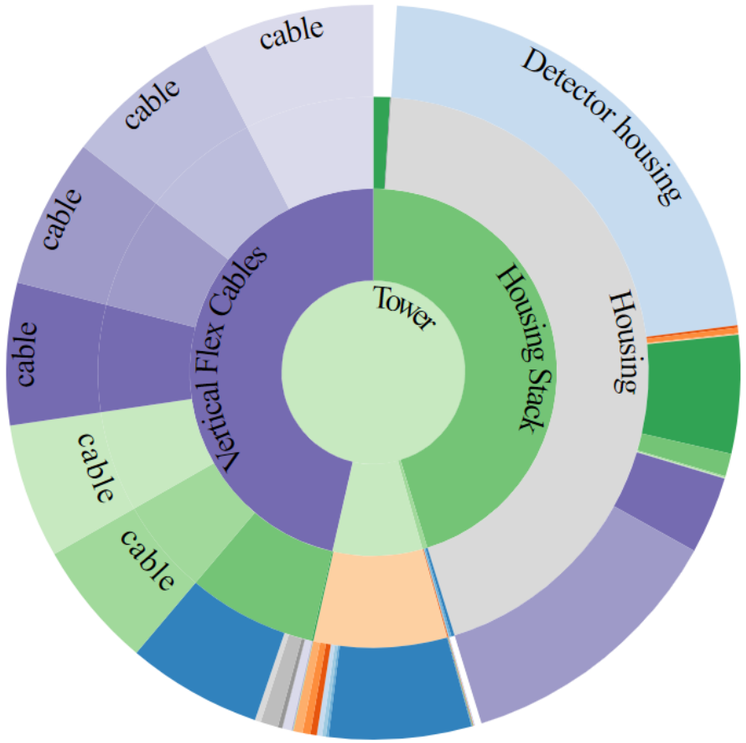
- Track component tree, assays, activation, radon, environment, and match to simulation results
- Import directly from radiopurity.org
- Total rates calculated with correlated, asymmetric errors and limits
- Branch, merge, and tag version control, configurable user permissions
- Easy one-line docker compose startup

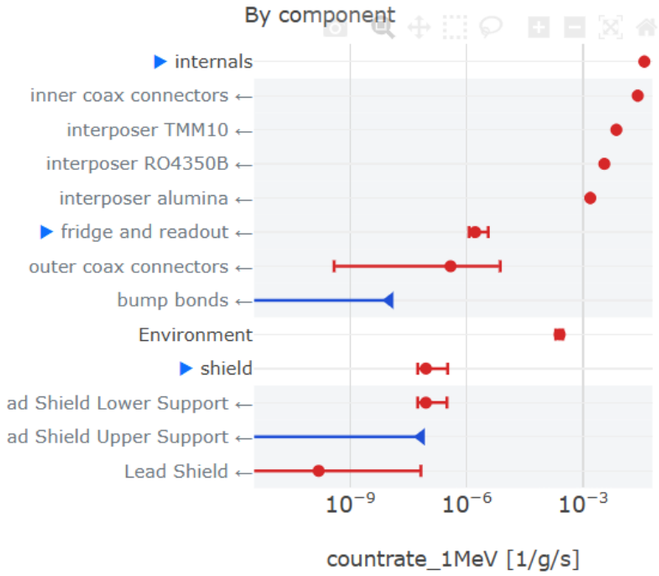


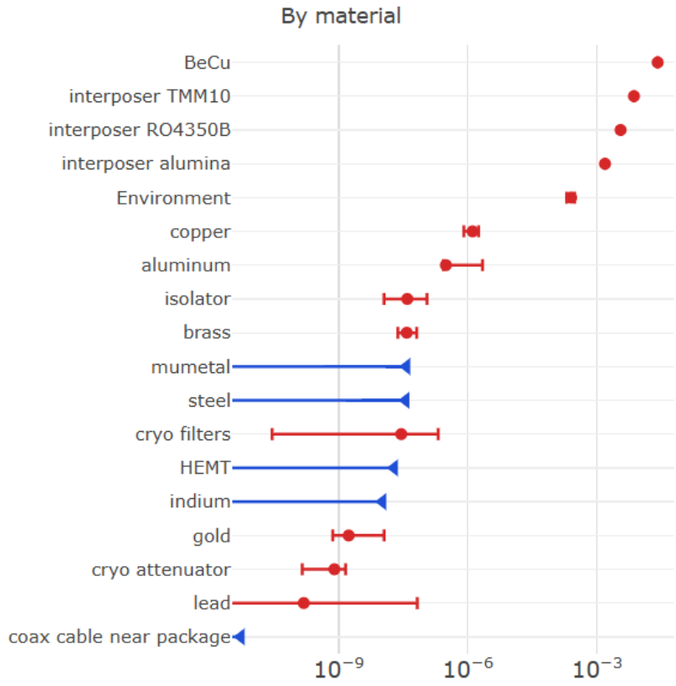


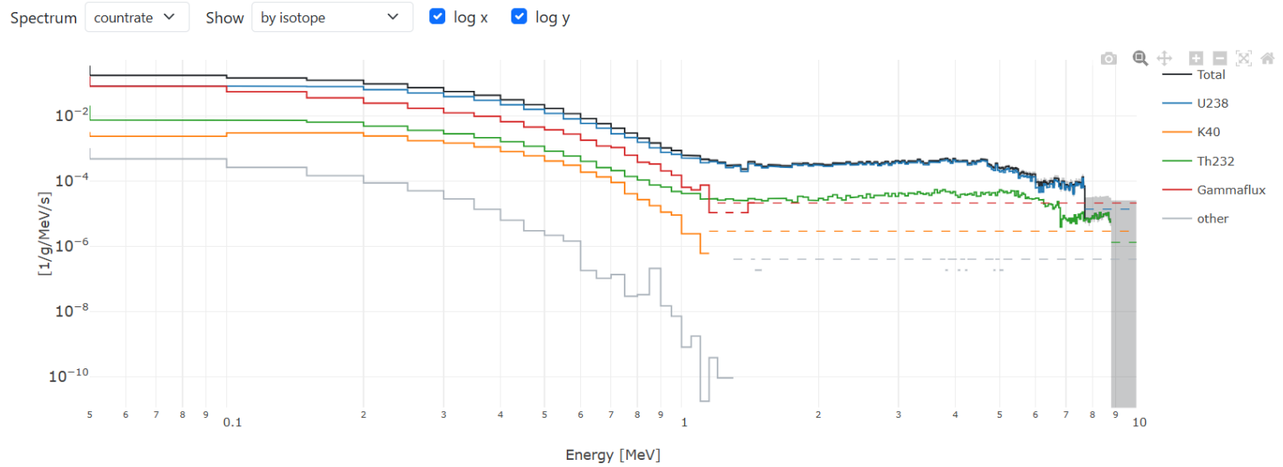
Background Modeling

Spectra and ROIs broken down by component, material, isotope, and category for easy visualization and analysis







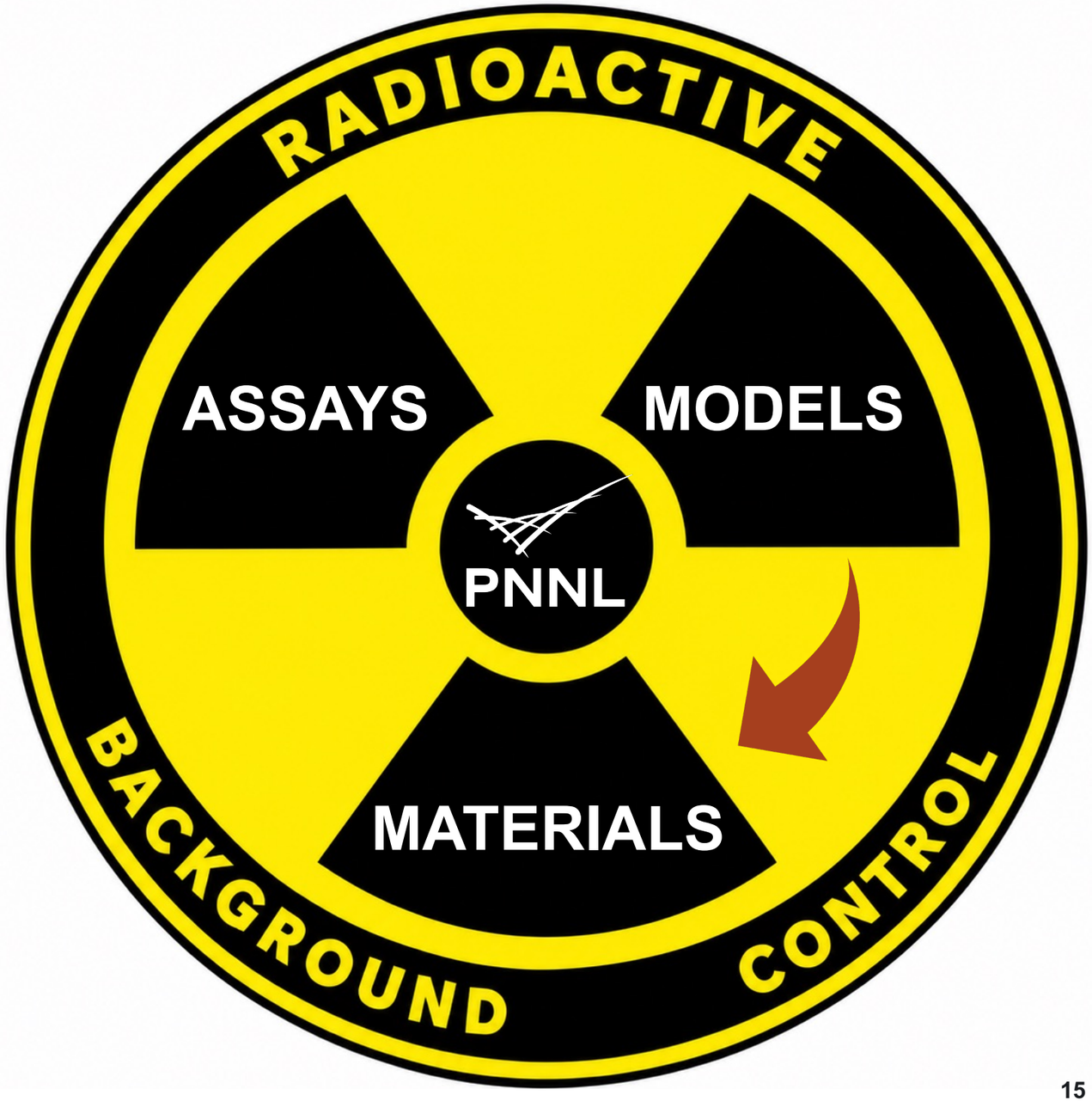


v2 at github.com/bloer/bgexplorer
v3 is in alpha and releasing soon

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What do you do when your experiment sensitivity is limited by radioactive backgrounds?

- Redesign detector layout
  - Use active and passive vetos
- Improve/develop analysis cuts to discriminate signals from backgrounds
- **Improve radiopurity of materials**

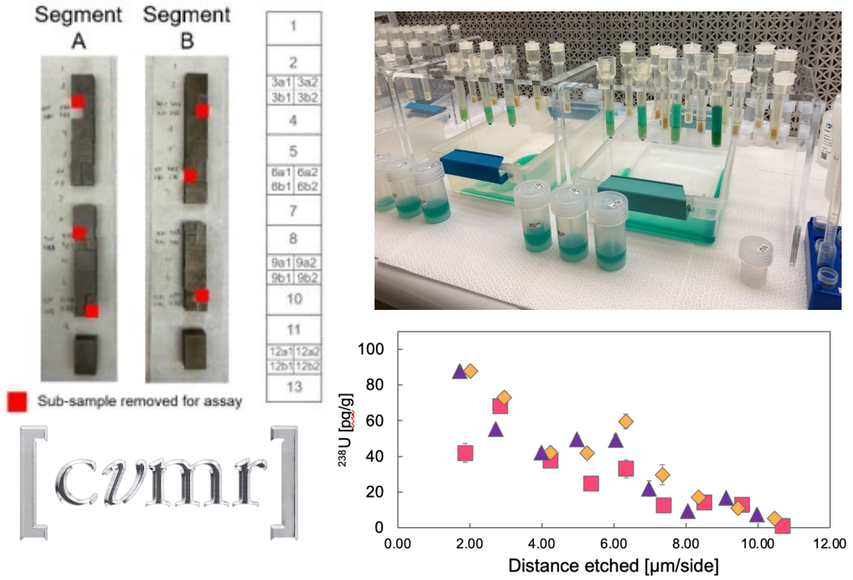


Developing Radiopure Components
CVD Metals

Chemical vapor deposition is a promising technique for producing extremely radiopure materials

Ultra-pure nickel via CVD

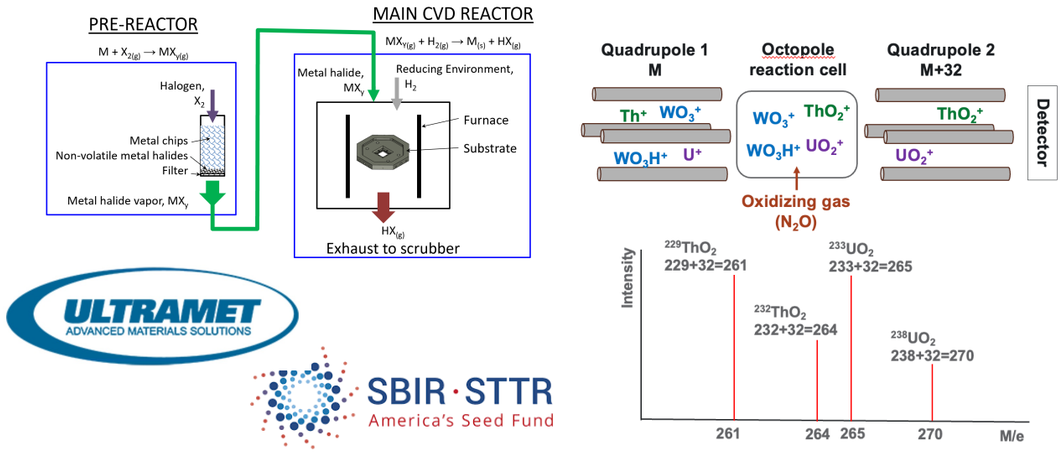
Depth profile of surface vs bulk contamination via chemical separation coupled with triple quadrupole mass spec.



Rosendaal T.J. et al. (nEXO Collaboration), NIM A 1087 (2026) 171402

Ultra-pure tungsten via CVD

Challenging material assay method via chemical separation coupled with triple quadrupole mass spec.



di Vacri M.L., Fortini A., Schlieder T.D., Hobbs K.P., Arnquist I.J., Hoppe E.W., and Huges D., NIM A 1082 (2026) 171018

Demonstrated impurities of ^{232}Th and ^{238}U in the material bulk in the ppq range (fractions of µBq/kg)

Developing Radiopure Components: Flexible Printed Cables

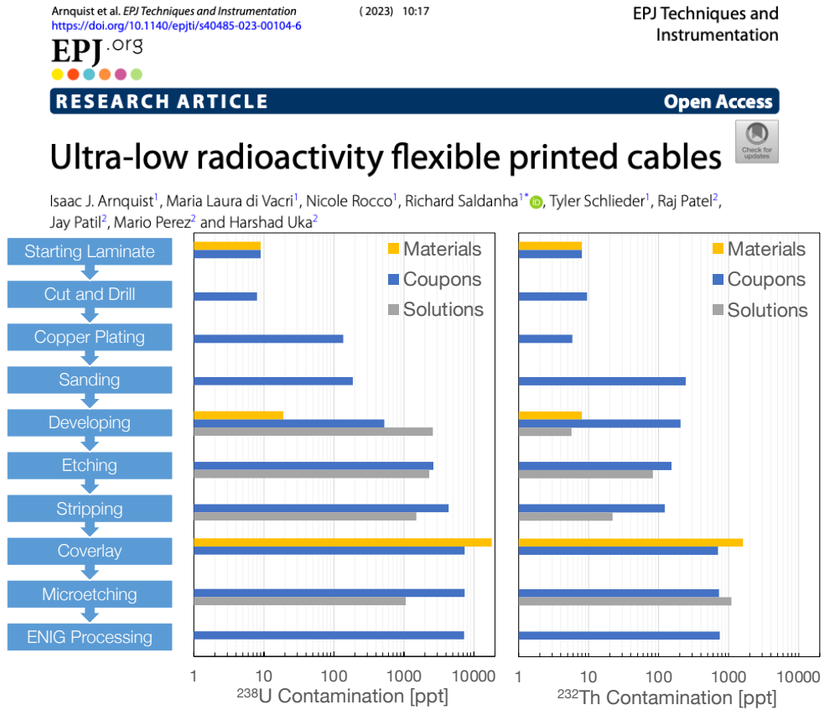
Worked with commercial company (through SBIR grant) to **investigate radioactive contamination in each individual fabrication step**

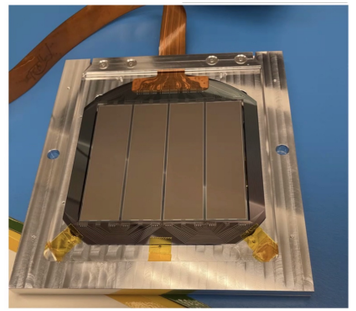
Developed **new radiopure fabrication process** that resulted in **5-100x reduction in U/Th/K contamination**

Cable	Copper layers [µm]	Polyimide layers [µm]	Coverlay	Surface finish	^{238}U [pg/g]	^{232}Th [pg/g]	^{nat}K [ng/g]
nEXO SiPM	**18 (x2)**	**50.8 (x1)**	**No**	**No**	**20 ± 2**	**<12.3**	**40 ± 12**
nEXO SiPM [Com.]	18 (x2)	50.8 (x1)	No	No	1300-6200	16-63	
DAMIC-M CCD	**18 (x2)**	**50.8 (x1)**	**x2**	**ENIG**	**31 ± 2**	**13 ± 3**	**550 ± 20**
DAMIC-M CCD [Com.]	18 (x2)	50.8 (x1)	x2	ENIG	2600 ± 40	261 ± 12	170 ± 50
EXO-200 [3, 12]	18 (x1)	25.4 (x1)	No	No	412 ± 47	<117	
EDELWEISS III [7, 14]	18 (x4)	25/125 (x3/x4)	No	No	650 ± 490	3700 ± 2500	2100 ± 840
DAMIC at SNOLAB [4]	18 (x5)	25.4 (x4)	x2	ENIG	4700 ± 400	790 ± 120	940 ± 60

Arnquist I.J., Beck C., di Vacri M.L., Harouaka K. and Saldanha R., NIM A 959 (2020) 163573

Arnquist I.J., di Vacri M.L., Rocco N., Saldanha R., Schlieder T.D., Patel R, Patil J., Perez M. and Uka H., EPJ Tech. and Instr. 10.1 17 (2023)





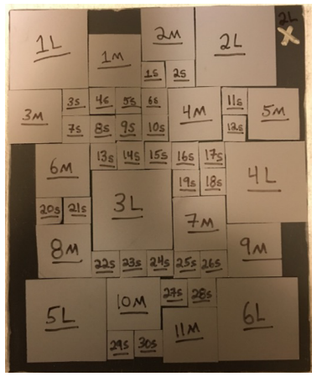


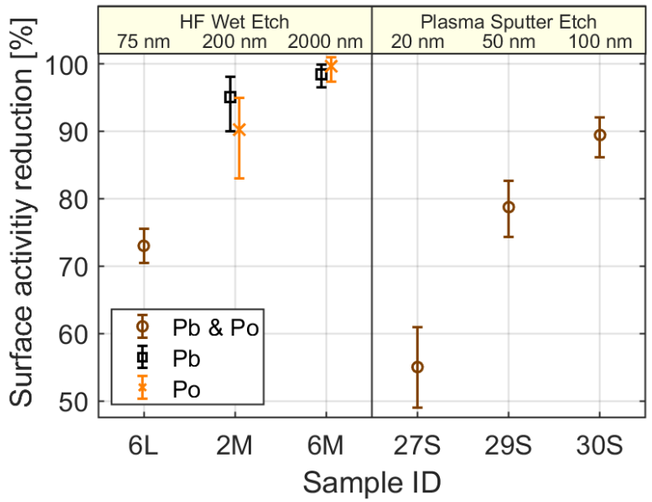
Delivered **28x 1.5-meter-long** flexible cables that met the radiopurity requirements for DAMIC-M.
10x reduction compared to off-the-shelf options
Now only ~3% of DAMIC-M background budget

Developing Radiopure Components: Material Surfaces

^{210}Pb removal from silicon surfaces

Explored ^{210}Pb removal using several different methods including washing, etching, baking, etc.

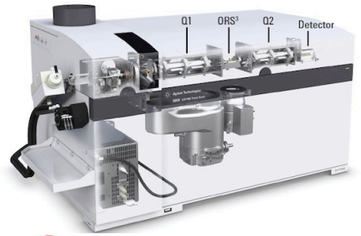


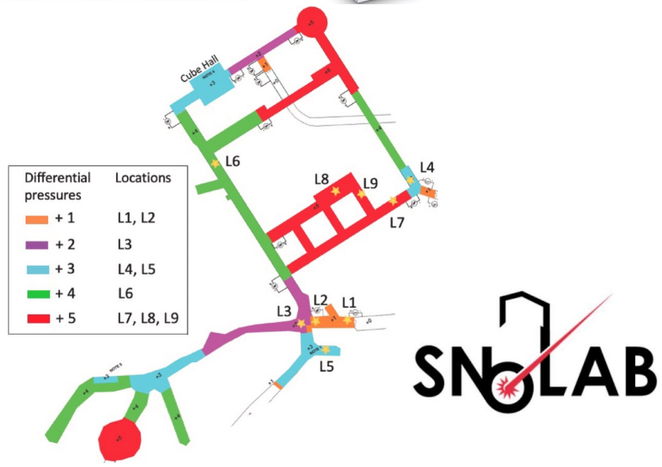


Arnquist et al., Exploration of methods to remove implanted ^{210}Pb and ^{210}Po contamination from silicon surfaces. *NIM A* **1048** (2023) 168008.

Determination of backgrounds from dust particulate





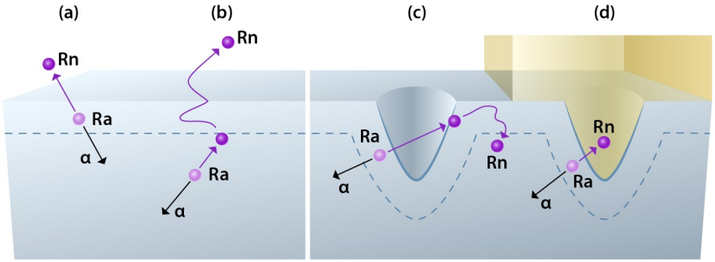


SNOLAB

di Vacri M.L., Arnquist I.J., Scorza S., Hoppe E.W. and Hall J., NIM A 994 (2021) 165051
di Vacri M.L., Scorza S., French A., Rocco N.D., Schlieder T.D., Hoppe E.W. and Hall J., NIM A 1056 (2023) 168700

Radon emanation and surface effects

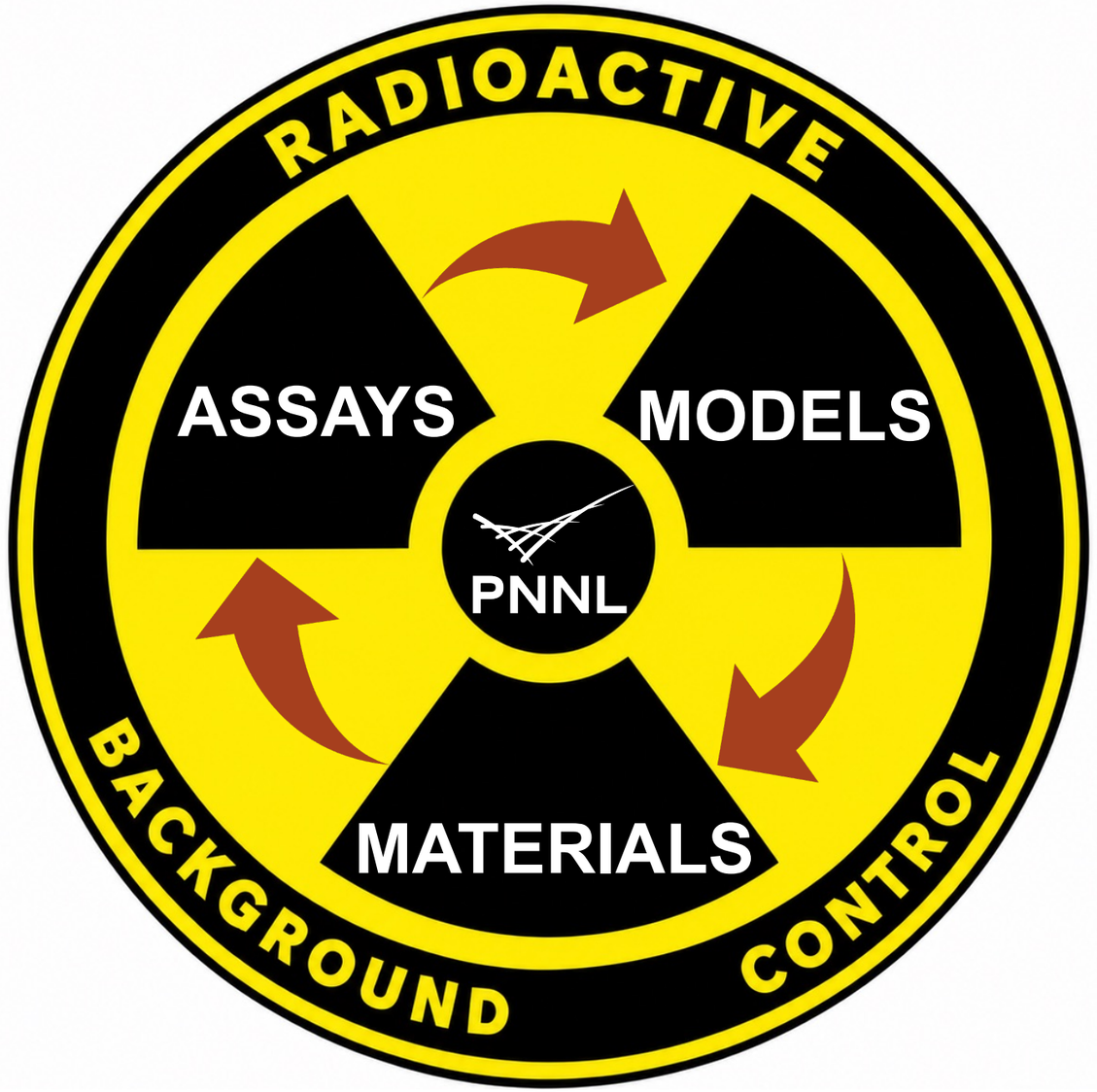
Modeled effects of surface roughness and environmental medium on radon emanation rates



Chris Jackson,
Tue Oct 20th, 5:27 PM
RDC 7
Low-Background Detectors

Cosmogenic Background Control

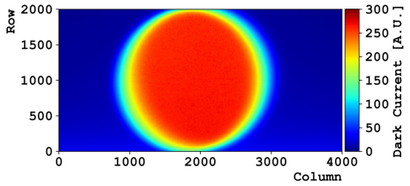
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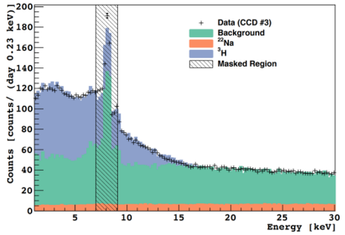


Tritium Production Rate

Neutron beam irradiation of CCDs

Self-counting of ^{3}H





First measurement of ^{3}H sea-level production rate:

124 atoms/kg/day

Phys. Rev. D 102, 102006 (2020)

Cosmogenic Exposure Models and Tracking

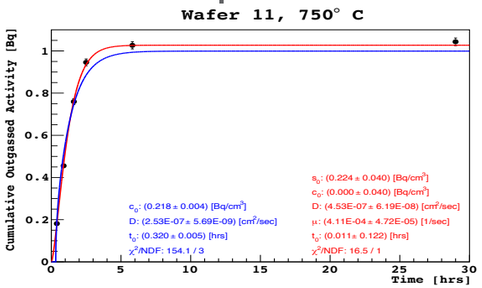
- Establishing **witness sample materials** for tracking exposure
- **Software package for cosmogenic activation** evaluation

Total background budget for OSCURA CCDs:

12 sea-level days

Tritium removal from Si

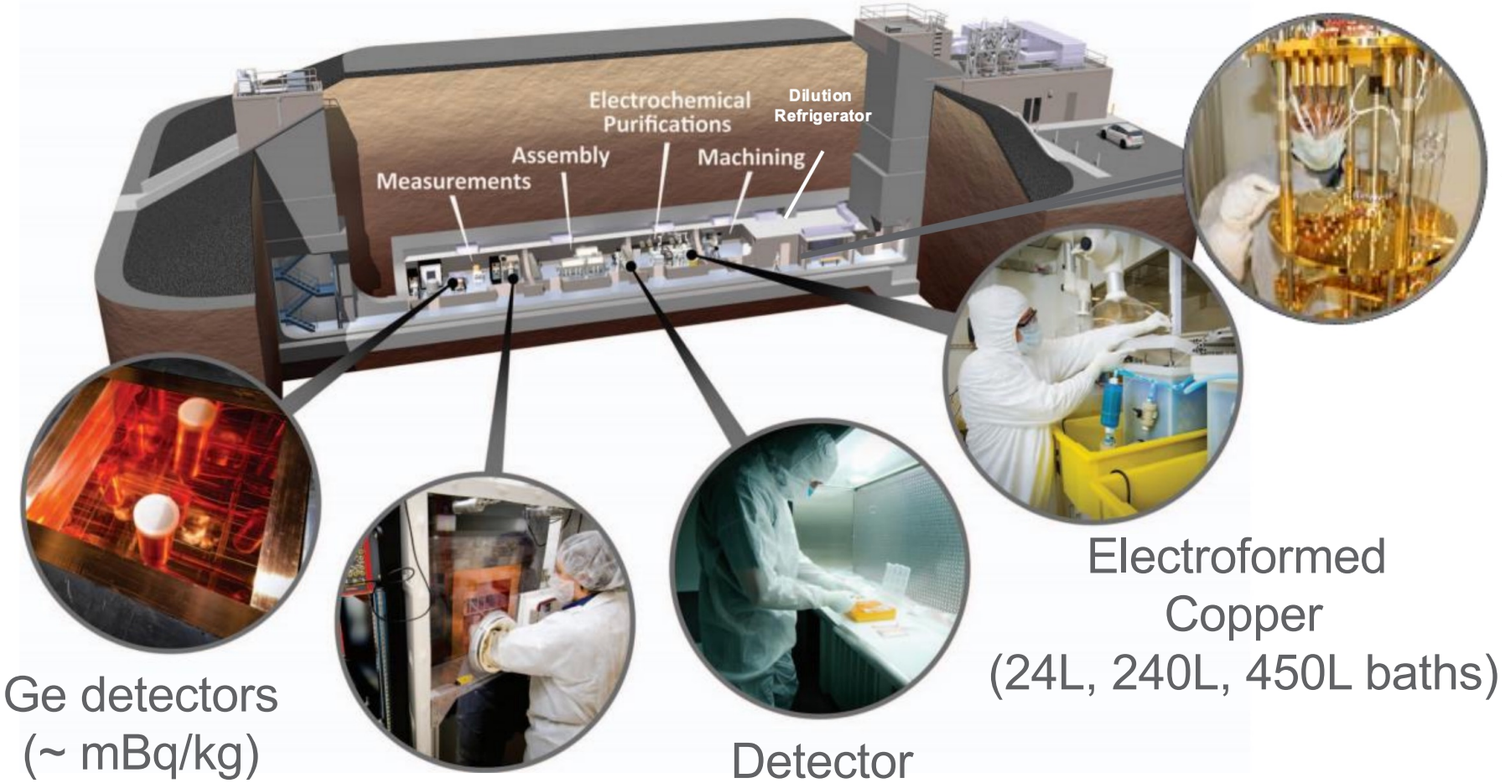
Demonstrated removal of cosmogenic tritium at elevated temperatures



> 95% removal from 725-um Si in 2.5 hrs @ 750 C

Phys. Rev. D 112, 052011 (2025)

Facilities:
PNNL Shallow Underground Laboratory (SUL)



Dilution Refrigerator (with Pb shield)
Liquid Scintillator Counter
XIA Surface Alpha Counter

Ge detectors
(~ mBq/kg)

Proportional Counters
(~ 25 mBq/kg ^{39}Ar)

Detector Assembly

Electroformed Copper
(24L, 240L, 450L baths)

19 m overburden, ~ 30 m.w.e.
Cosmic flux Reduction:
Muon flux: 6x
Neutron and proton flux: >100x

~ 6600 sq. ft.

Clean rooms
(class 10,000 and 1,000)

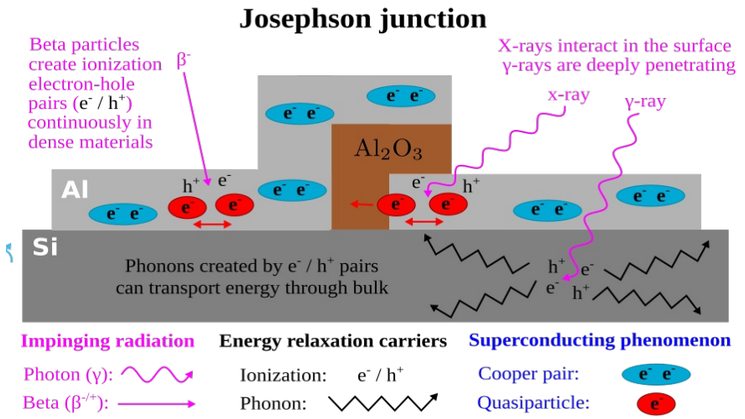
Shallow underground laboratory at PNNL supports ultra-sensitive radiation detection for both fundamental physics and applied national security mission

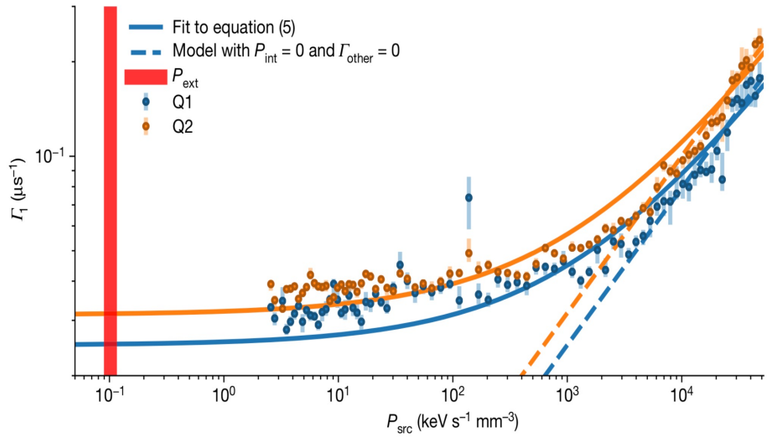
Radiation and Quantum Devices

Demonstrated degradation of superconducting qubit performance when exposed to radiation

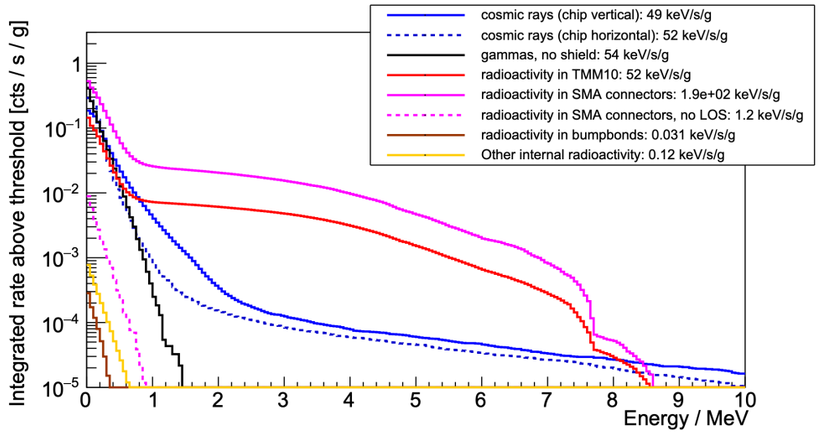
Based on systematic assays and modeling of silicon chip substrates in a typical dilution refrigerator setup, identified various potential sources of external and internal radiation that can affect performance

Recently completed study on multi-MeV energy depositions as a source of correlated events:
Samuel Watkins,
Tue Oct 20th, 5:47 PM
RDC 7
Low-Background Detectors

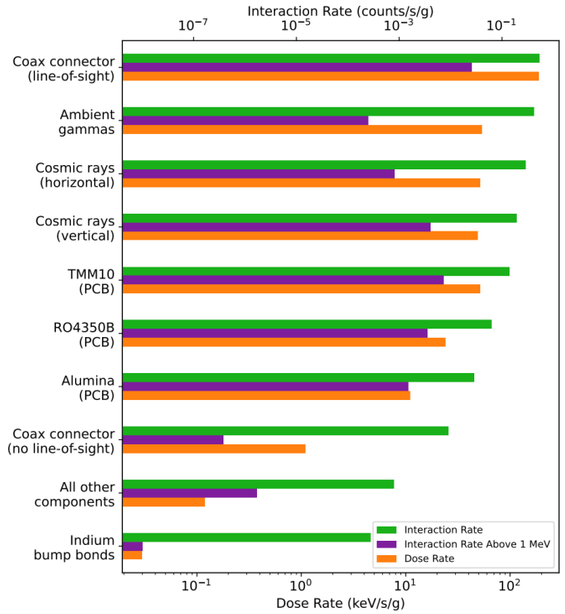




Vepsalainen et. al. Nature **584**, 551–556 (2020)



Loer et al., JINST 19 P09001 (2024)





Summary

- Pacific Northwest National Laboratory (PNNL) has a long history of R&D in radioactive background control
- PNNL hosts an ultra-sensitive ICP-MS assay facility with world-leading sensitivities to radioactive contaminants
- Custom developed low-radioactivity materials and components have been utilized in a wide range of rare event searches
- Software packages for background modeling, cosmogenic activation, and radon emanation estimations have been developed
- **If you have concerns about radioactive backgrounds in your experiment, please reach out: richard.saldanha@pnnl.gov**

Pacific Northwest
NATIONAL LABORATORY

Thank you



