Ultra-pure Copper Electroforming for Background Suppression in Rare-Event Searches

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Copper as a Construction Material

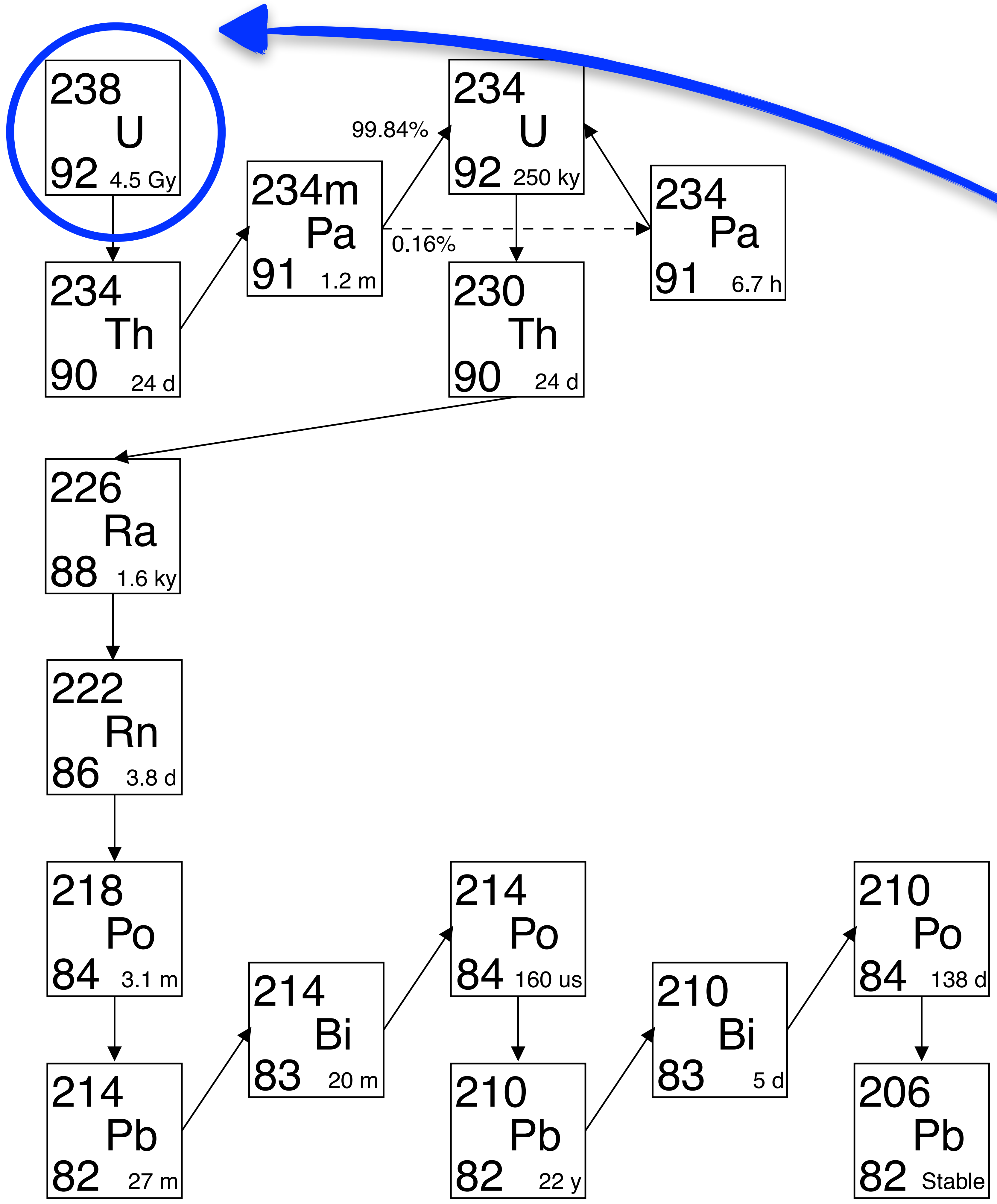
- Copper is common material for rare event searches
  - Strong enough to build limited-pressure vessels or support structures
  - Commercially available at high purity
  - Low cost
  - No long-lived radio-isotopes
    - Longest is ^{67}Cu, $t_{1/2}$= 62 hours

Radioactive Contaminants in Copper

Commercial copper has two primary contamination sources:

- Fast neutrons from cosmic muon spallation e.g. $^{63}Cu(n,\alpha)^{60}Co$
  - Mitigation: minimise time outside underground laboratory

Radioactive Contaminants in Copper



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  - Assay: ICP-MS ~10 µBq/kg

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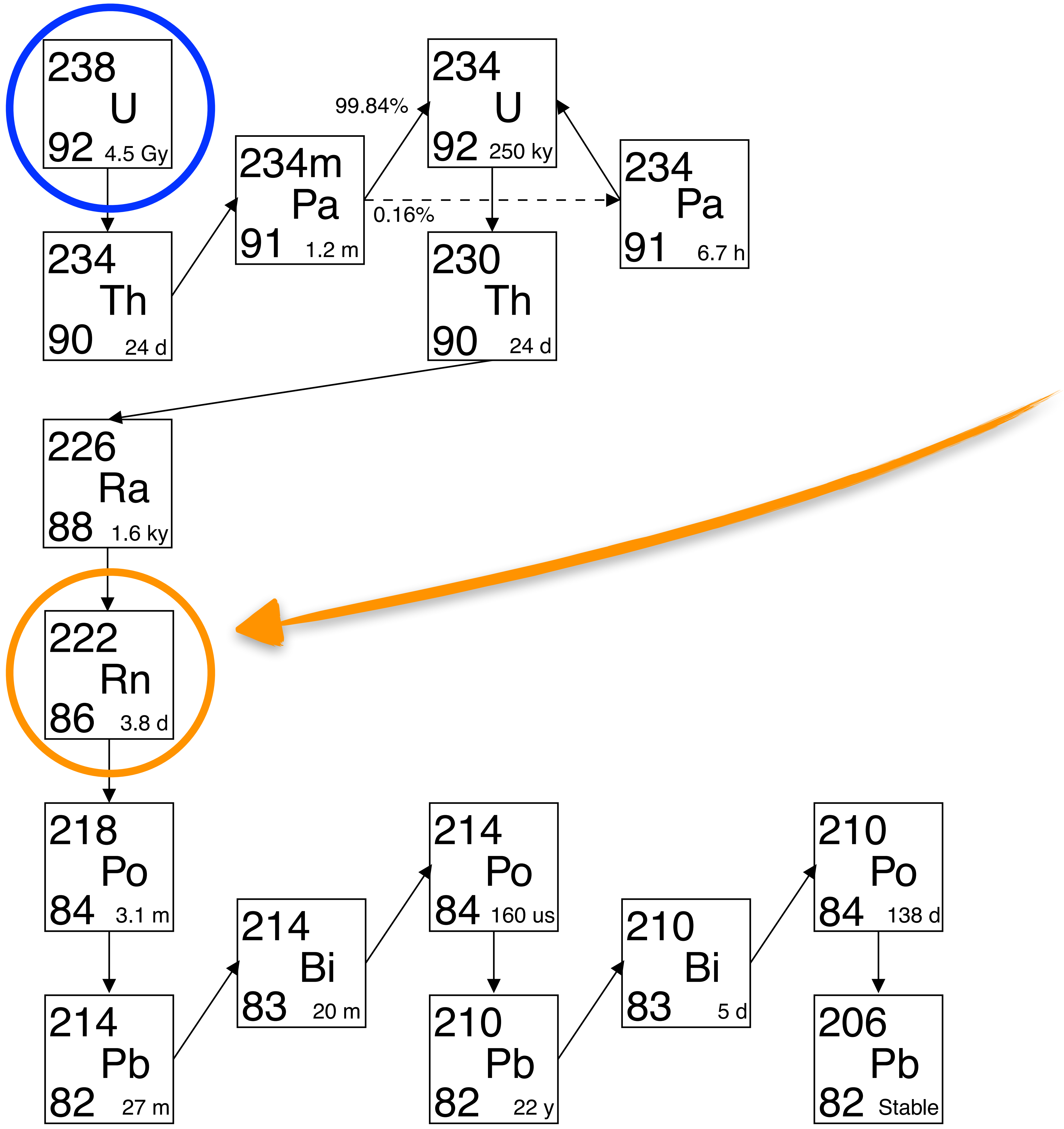

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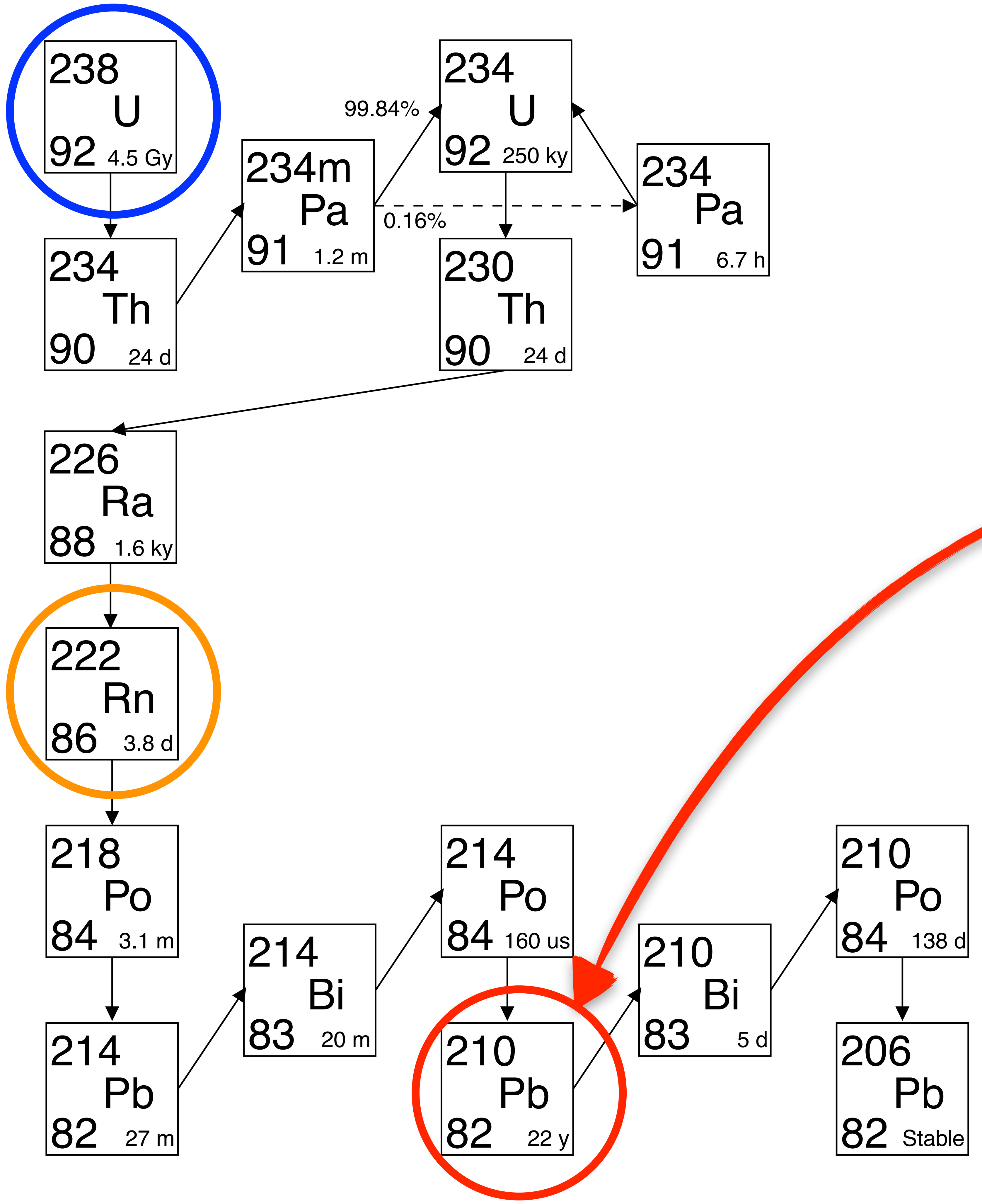

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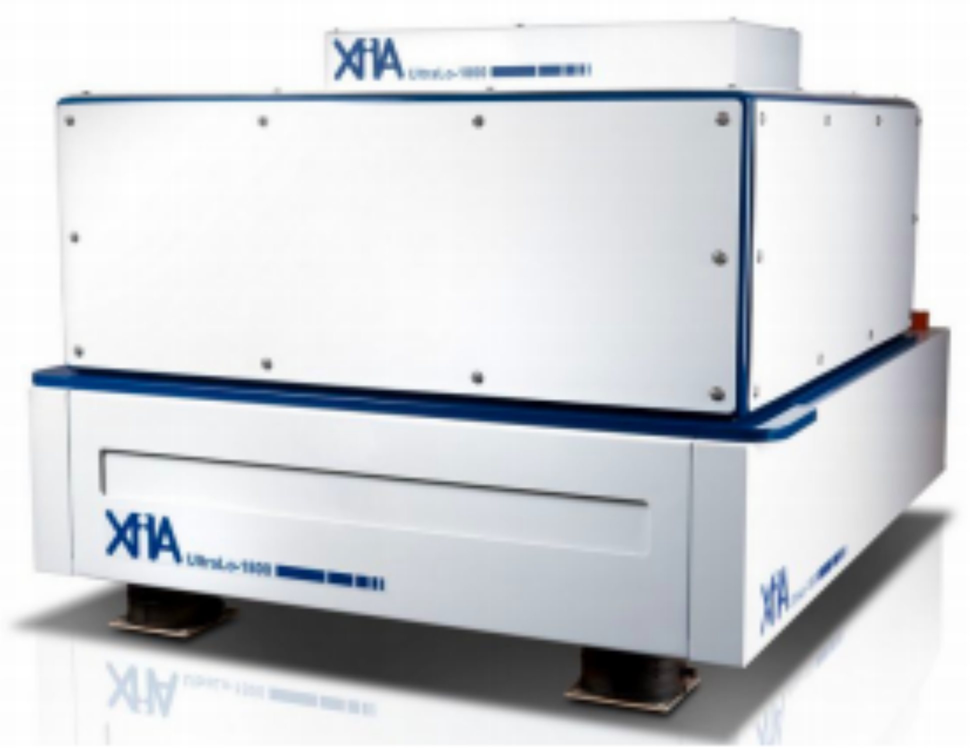
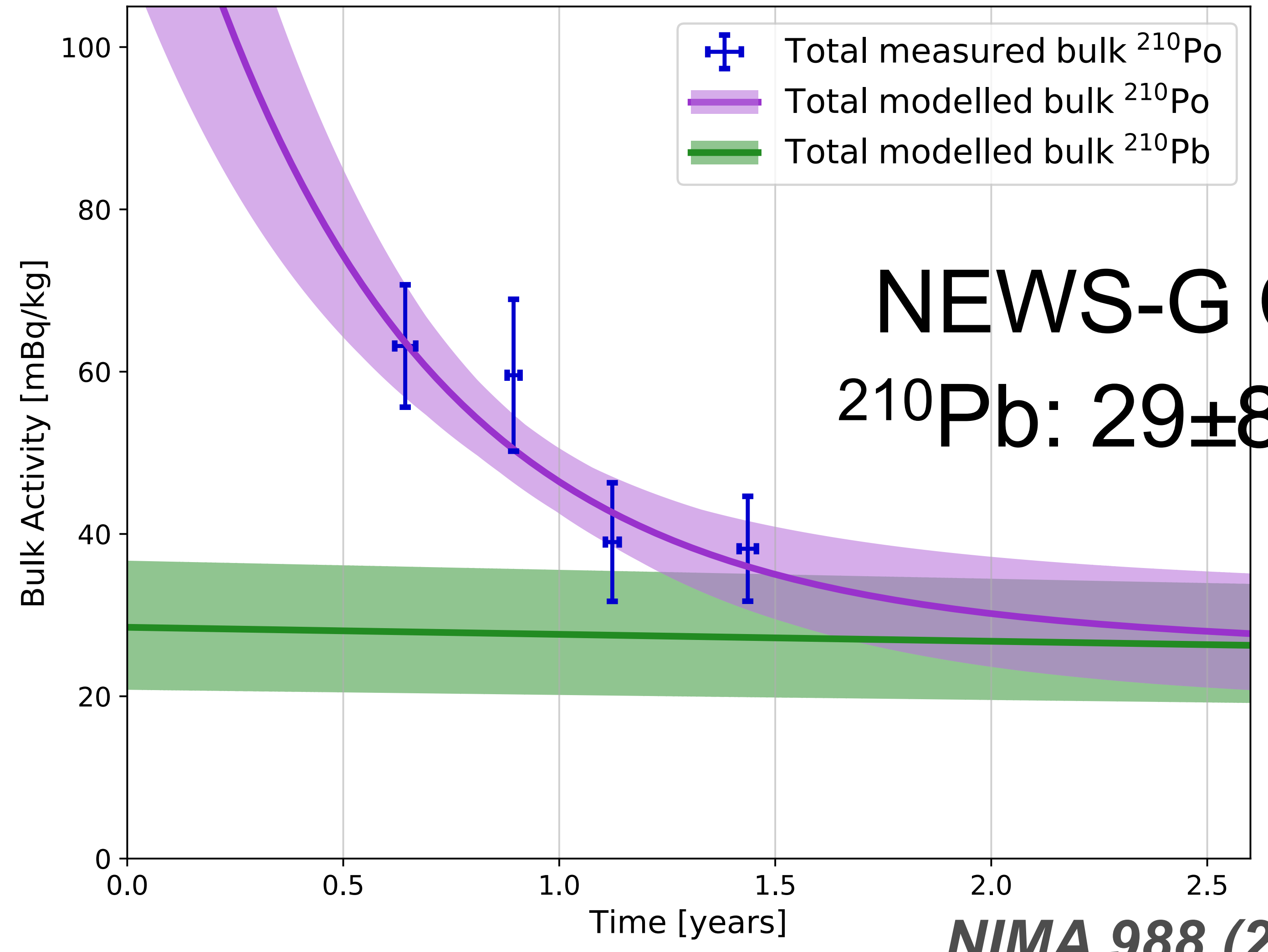
  - Mitigation: minimise time outside underground laboratory
- **^{238}U and ^{232}Th** decay chains naturally found in raw material
  - Assay: ICP-MS ~10 μBq/kg
- Long-lived **^{238}U** daughters introduced by **^{222}Rn gas**
  - **^{210}Pb is long-lived**, so builds up, and leads to **break in secular equilibrium** of chain
  - Assay: alpha-counter, UltraLo-1800 ~30 mBq/kg



XIA UltraLo-1800

https://www.xia.com/ultralo-theory.html



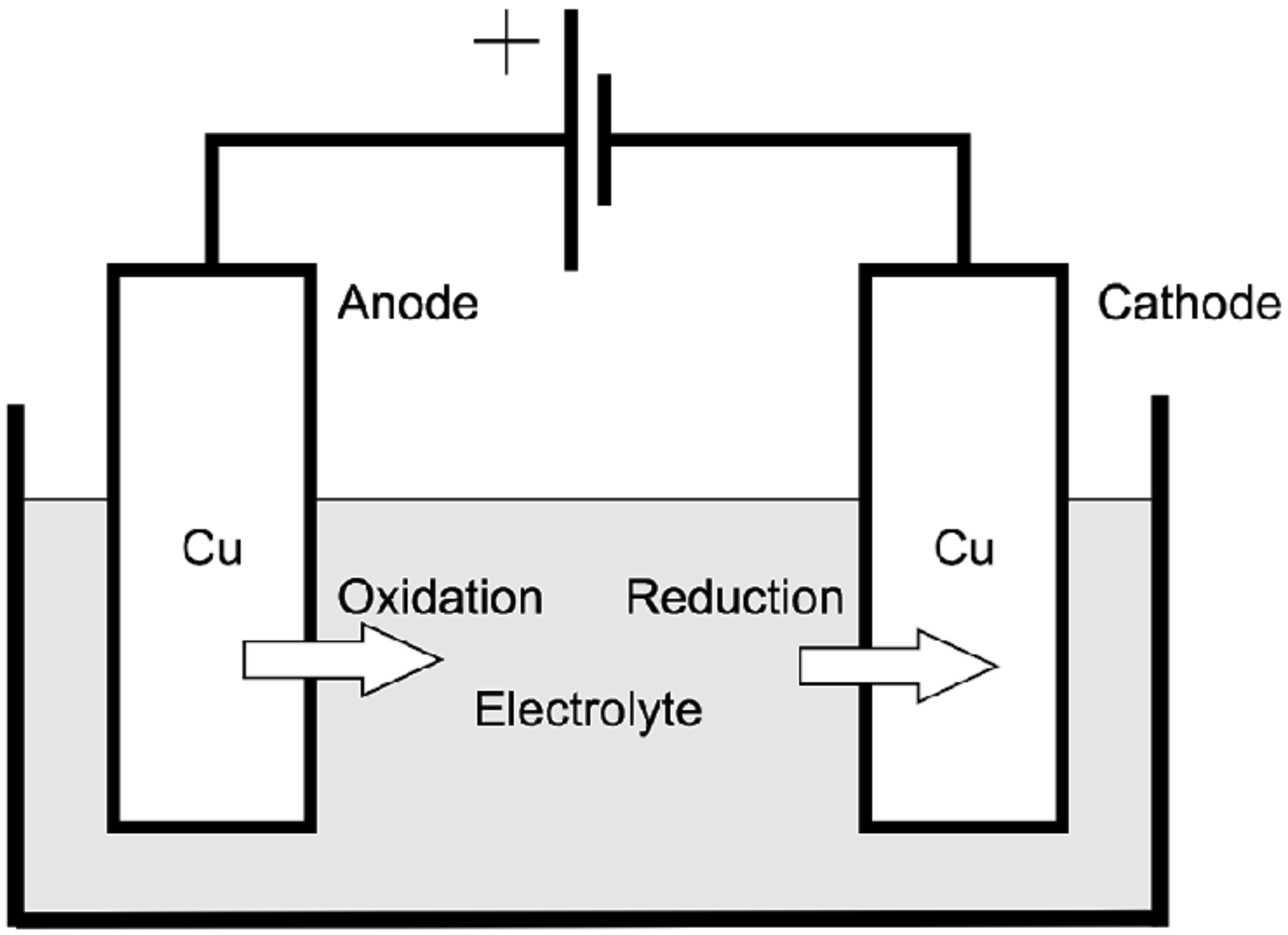
NEWS-G Copper:
^{210}Pb: 29±8 mBq/kg

NIMA 988 (2021) 164844

Electrolytic Cell and Electroplating

- **Electrolysis** governed by oxidation and reduction reactions
  - Reduction of ions requires supply of electrons → Current
  - Also requires energy → Potential difference
- **Electroforming:** Ions gain electrons and deposit as atoms (reduction) at cathode → material build-up
  - Supplied current drives reaction
  - Deposited mass proportional to current:

$$M = \frac{m_r \int I(t) \mathrm{d}t}{zF}$$



M - mass deposited

m_r - molar mass

$I(t)$ - current

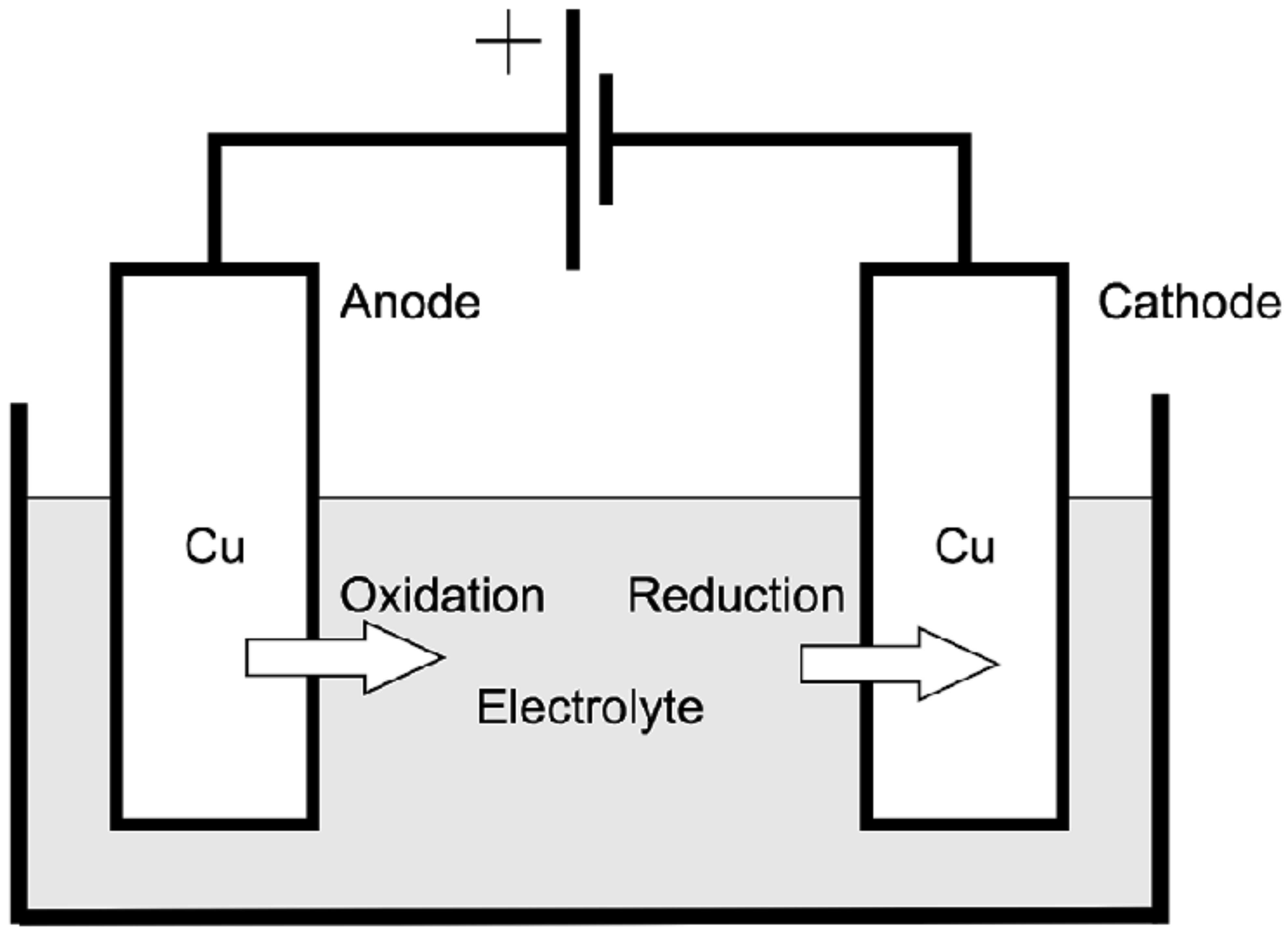
z - number of electrons transferred

F - Faraday Constant($= eN_A$)

Purification Through Electroplating

- Some ions reduce more readily than others
  - **Reduction potentials**
- Copper benefits from 'electrowinning' - high reduction potential +0.34 V
- Contaminants have lower reduction potential than copper
  - Copper refined during electroforming if electrode potential is low enough

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



Copper Purification

- Reaction that proceeds determined by standard cell potential:

$$E^0_{cell} = E^0_C - E^0_A$$

- Related to change in Gibbs Free Energy:

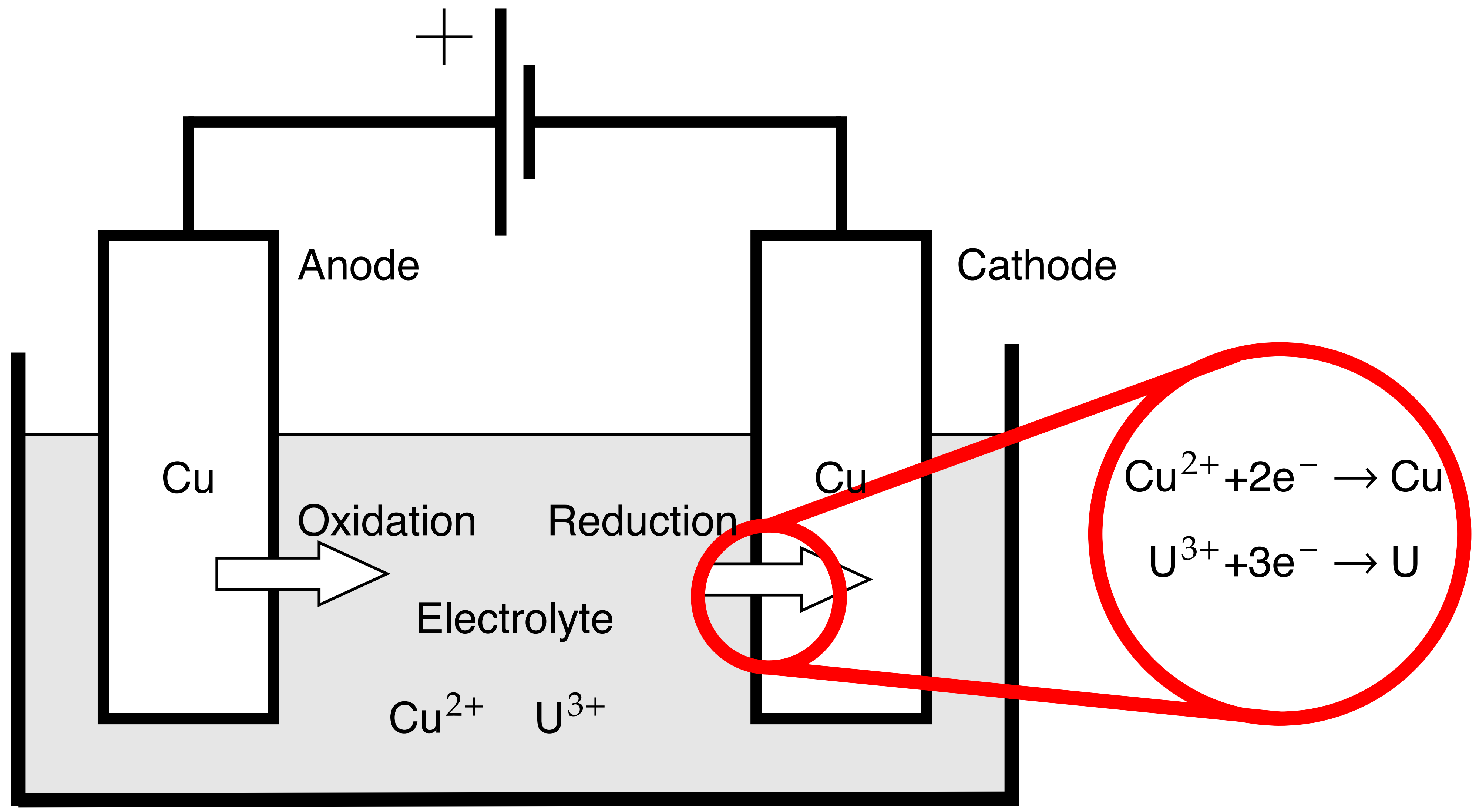
$$\Delta G^0 = -zFE^0_{cell}$$

- If $\Delta G^0 < 0$, then reaction is spontaneous
- If $\Delta G^0 > 0$, then extra energy is needed

E^0_{cell} - standard cell potential
E^0_C - standard reduction potential at cathode
E^0_A - standard reduction potential at anode

Example of electrolyte containing U^{3+} and Cu^{2+} ions, with a Cu anode:

- To reduce U^{3+} to U:
  - E^0_{cell} = -2.14 V → Requires energy
- To reduce Cu^{2+} to Cu
  - E^0_{cell} = 0 V → In equilibrium
- Cu^{2+} reduction is energetically favourable to U^{3+} reduction
- Potential difference required to drive reaction and overcome energy losses

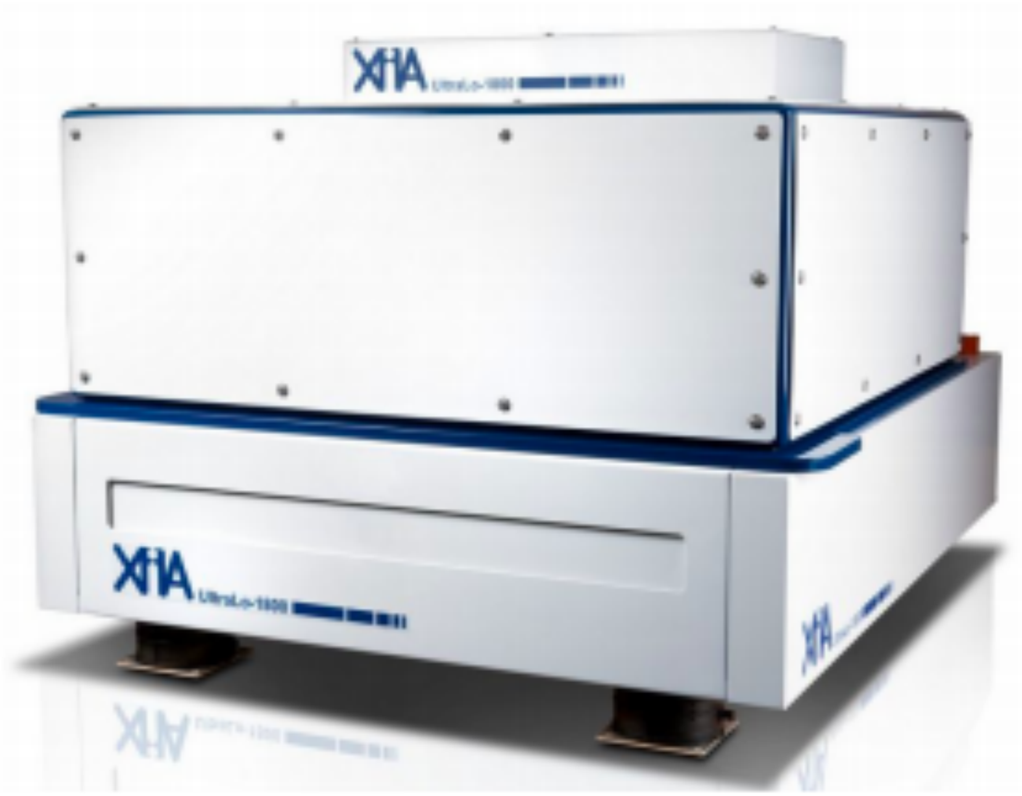


Radiopurity of Electroformed Copper

- Current ^{238}U and ^{232}Th contaminations below sensitivity of most sensitive assay technique - ICP-MS
  - Bounds are just upper limit – value may be much lower
- ^{210}Pb assayed with XIA UltraLo-1800 α-particle counter
  - Again, below sensitivity of device

ICP-MS Assay

Copper Type	^{232}Th [ppt] (*μBq/kg*)	^{238}U [ppt] (*μBq/kg*)	Source
C10100	0.46±0.06 (*1.19*±0.25)	0.21±0.06 (*2.54*±0.74)	Majorana Demonstrator, (PNNL) 10.1016/j.nima.2016.04.070
Electroformed	<0.029 (<*0.11*)	<0.008 (<*0.10*)	Majorana Demonstrator, (PNNL) 10.1016/j.nima.2016.04.070
Electroformed	0.035±0.004 (*0.14*)	<0.050 (<*0.06*)	CES, LSC 10.1063/1.5018987



XIA UltraLo-1800
https://www.xia.com/ultralo-theory.html

XIA UltraLo-1800 Assay

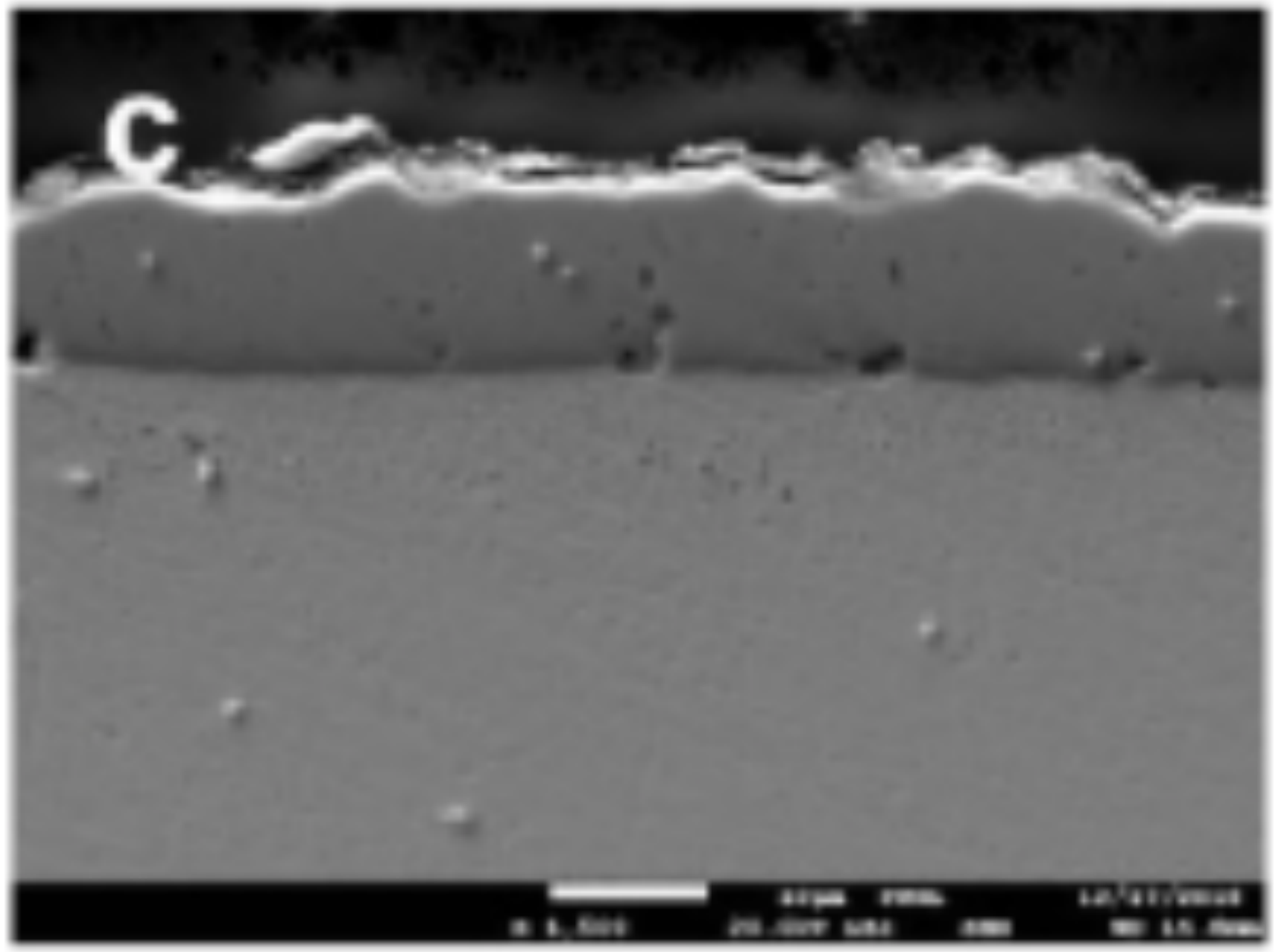
Sample	^{210}Pb contamination (mBq/kg)	^{210}Po contamination (mBq/kg)
OFC#1 (C1020) (MMC)	40±8	47±21
OFC#2 (C1020) (MMC)	20±6	33±14
OFC#3 (C1020) (MMC)	27±7	(1.6±0.3)x10^2
OFC#4 (C1020) (MMC)	23±8	(2.2±0.4)x10^2
OFC#5 (C1020) (SH copper products)	17±6	44±18
OFC#6 (C1020) (SH copper products)	27±8	24±17
OFC (class1) (SH copper products)	36±13	38±3
Coarse copper (MMC)	(57±1)x10^3	(16±2)x10^3
Bare copper (MMC)	8.4±4.0	(1.1±0.2)x10^2
OFC (MMC)	23±8	(1.3±0.3)x10^2
6N copper (MMC)	<4.1	<4.8
Electroformed copper (Asahi-Kinzoku)	<5.3	<18

NIMA 884 (2018) 157-161

PureAlloys

- Copper has drawback of low strength and high ductility
- Alloys generally used to improve mechanical properties
- Recent exploration of ultra-pure Cu-Cr layered alloys at PNNL
  - Alternately deposit Cu and Cr, and heat to alloy
  - Benefit from improved strength
  - Work ongoing to identify minimum Cr to keep radioisotopes low
  - 50% increase in hardness with 1% (w/w) Cr
- Would allow less material to be used, speeding up process
- Birmingham exploring collaboration with material scientists to study and model the processes

Cr electroplated on EFCu



NIMA 1003 (2021) 165291

Use by Rare Event Searches

- Ultra-pure copper electroforming already used by several experiments
  - Majorana Demonstratoe — NIMA 828 (2016) 22-36
  - NEWS-G — NIMA 988 (2021) 164844
  - ANAIS — Eur.Phys.J.C 79 (2019) 3, 228
- Mentioned as a prerequisite for future experiments
  - nEXO — 1805.11142
  - NEXT — JINST 13 (2018) 12, P12010
  - LEGEND — 1905.06572
  - …

MAJORANA Demonstrator,

Cryostat IR Shield

https://www.npl.washington.edu/majorana/majorana-experiment

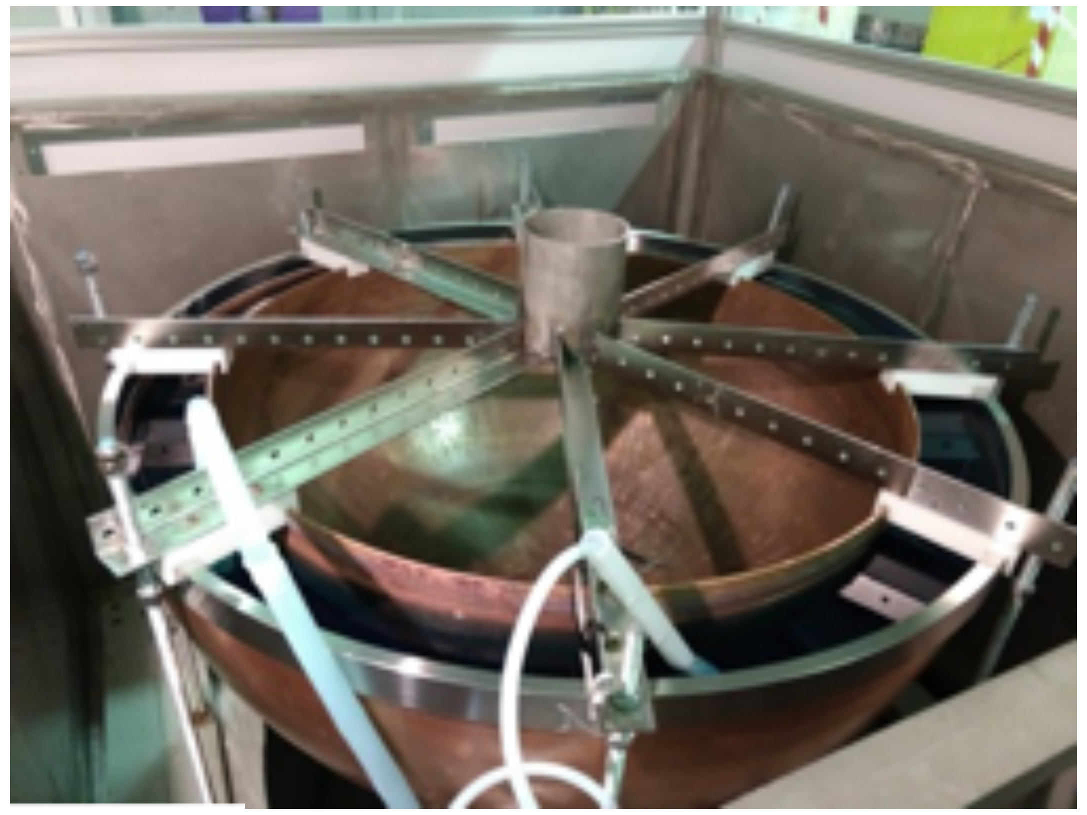
NEWS-G

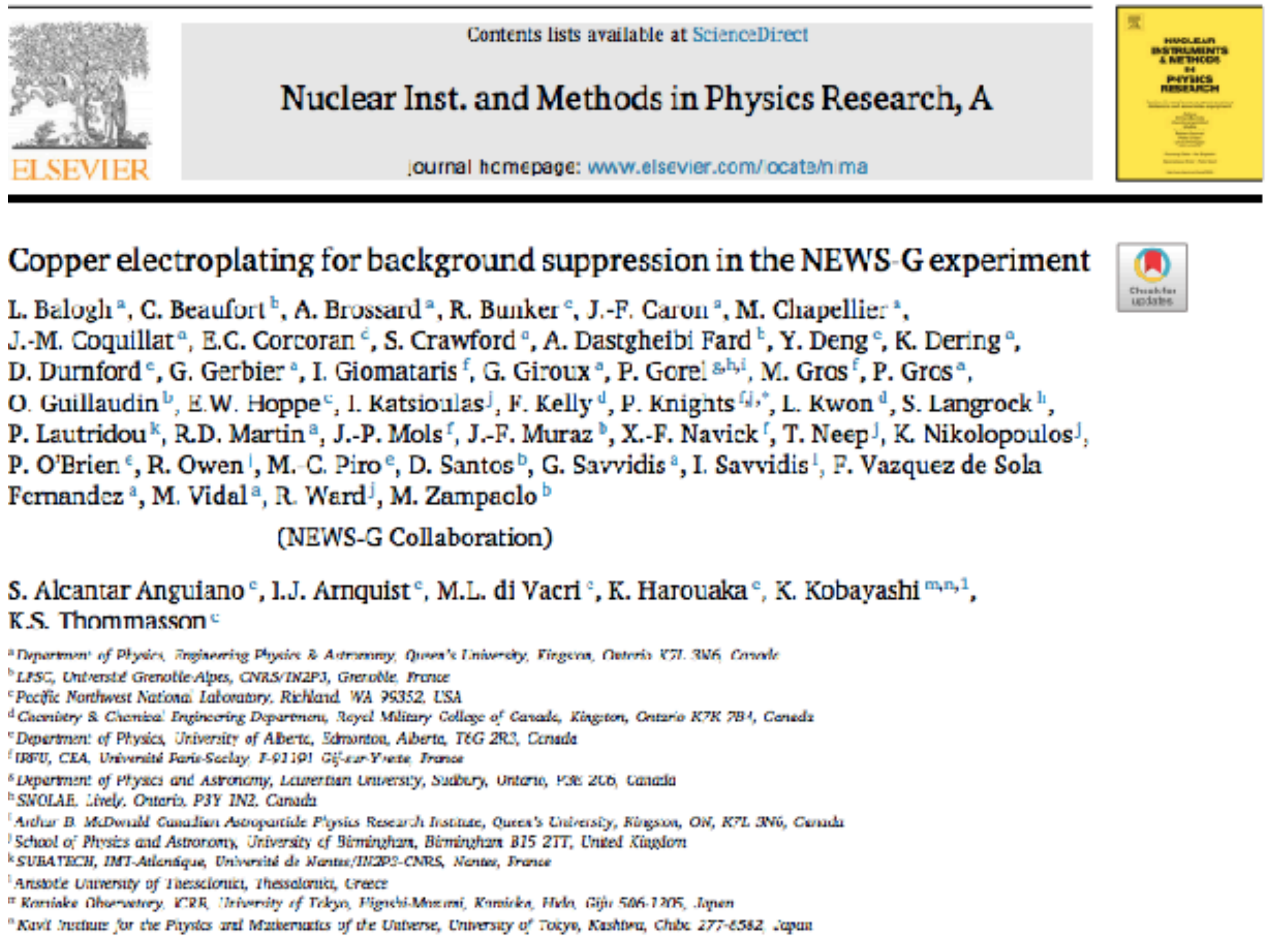
Electroplating

NIMA 988 (2021) 164844

Deep Underground Electroformed Copper

- Electroforming underground suppresses background contribution from cosmogenic activation
- Already demonstrated feasibility by NEWS-G collaboration
  - 500 μm layer plated to inner surface of 140 cm detector
  - Plating rate ~1 mm/month





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Copper electroplating for background suppression in the NEWS-G experiment

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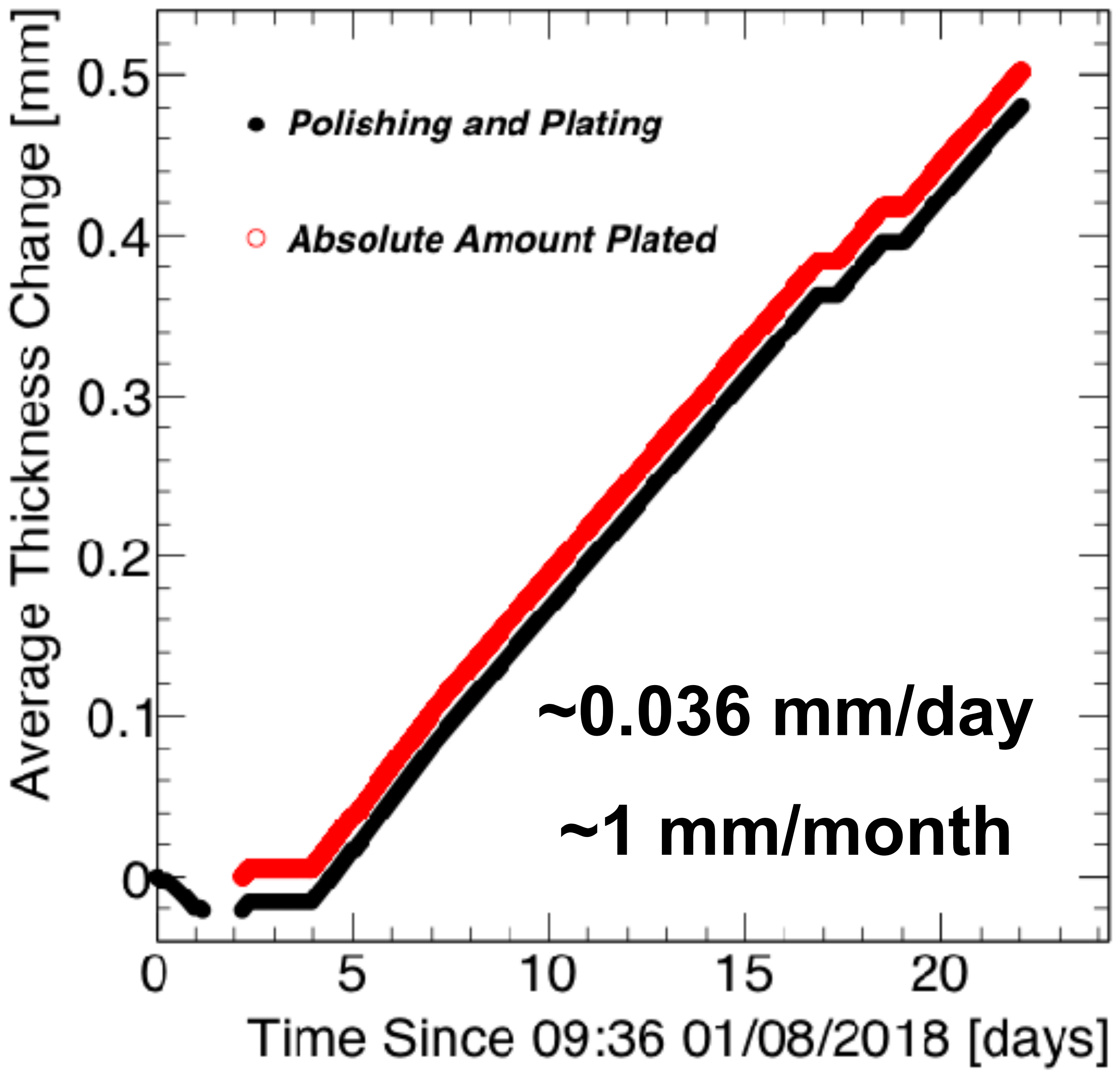
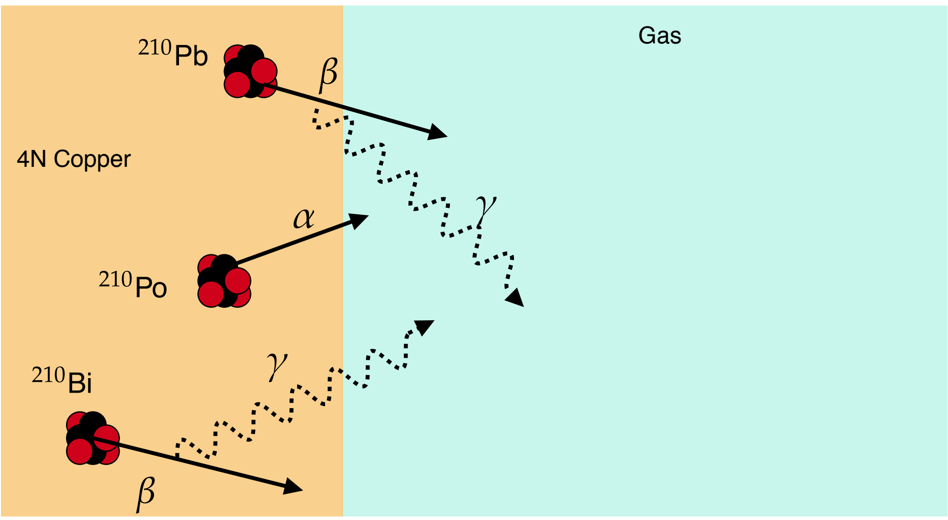
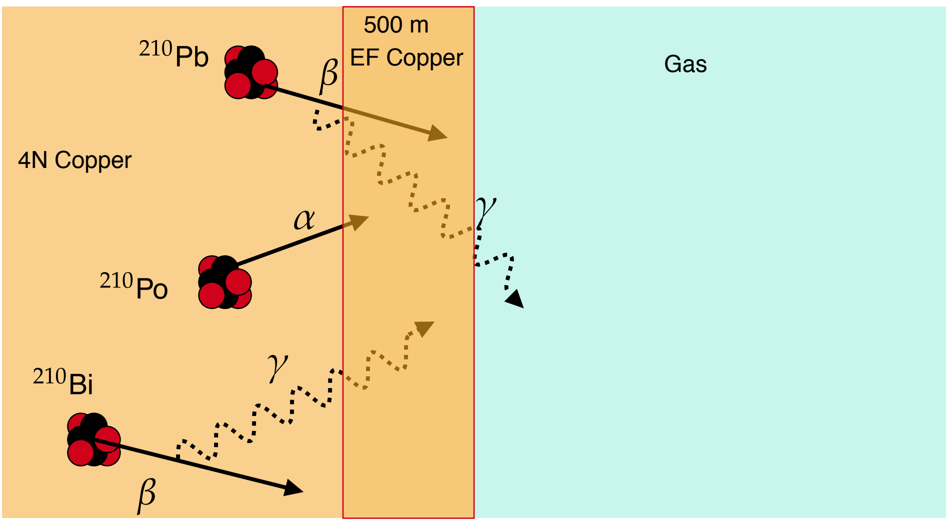
NIMA 988 (2021) 164844







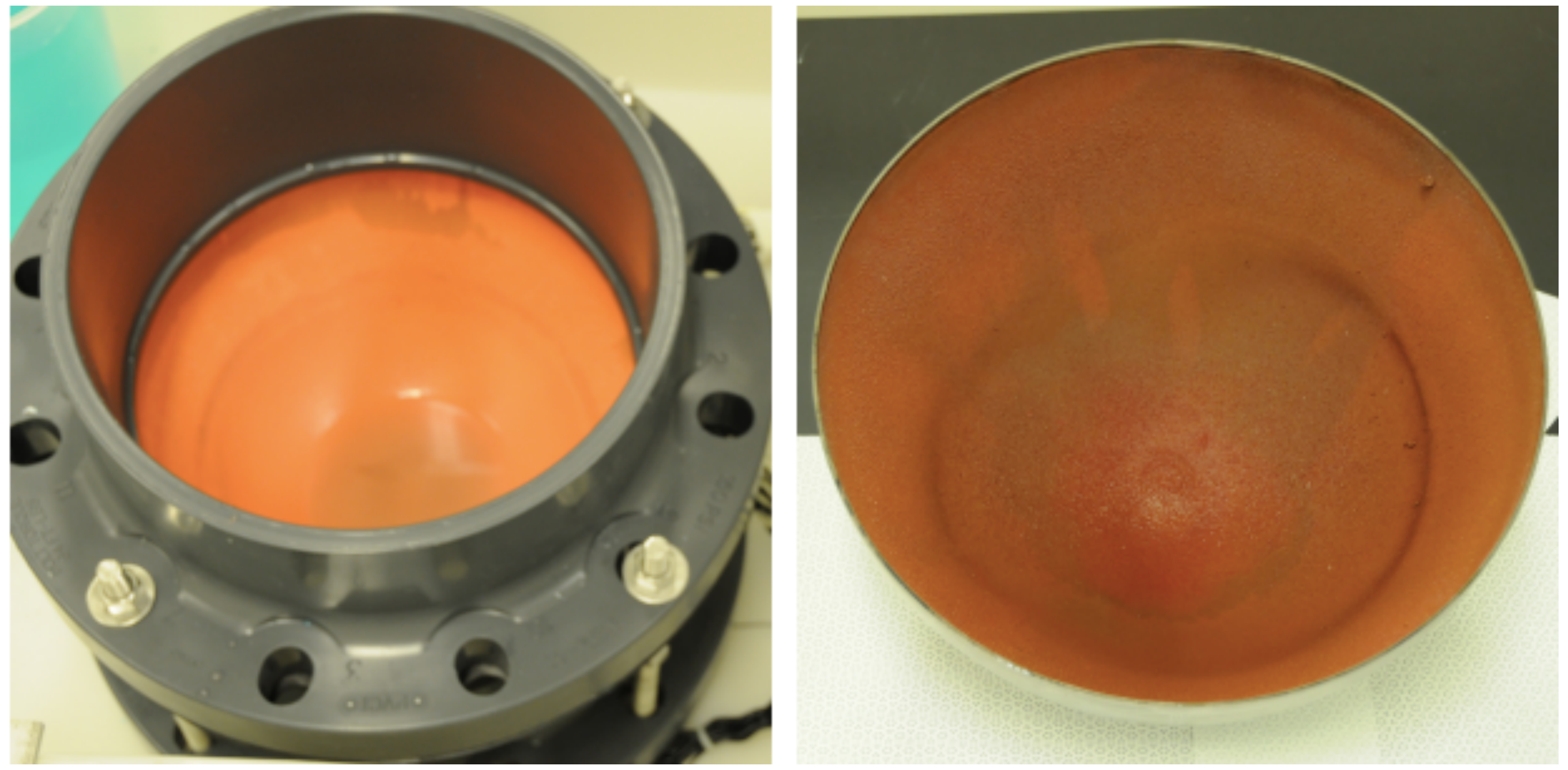
Sample	Weight [g]	^{232}Th [μBq/kg]	^{238}U [μBq/kg]
C10100 Cu (Machined)	-	8.7 ± 1.6	27.9 ± 1.9
Cu Electroformed	-	< 0.119	< 0.099
Hemisphere 1	0.256	< 0.58	< 0.26
Hemisphere 2	0.614	< 0.24	< 0.11

ECuME in SNOLAB

- 1.4 m diameter SPC electroforming - ~2 m diam electroforming bath
- Largest underground ultra-pure Cu electroforming facility
- Leadership from PNNL, and input from SNOLAB, Queen’s U. (Canada), Birmingham
  - Strong Birmingham involvement in project, and in previous electroplating
- Initially dedicated for an SPC, then will be available for users

Status:

- R&D for 30 cm prototype bath at PNNL
  - Procurement of parts underway
- Prototype to begin electroforming soon
- Full-scale planned to be underway early 2022
- Physic exploitation will follow SNOGLOBE



Prototype for NEWS-G Electroforming

Electroforming in Boulby

- STFC awarded funding for ‘scope, feasibility and costing’ study of establishing an electroplating facility in Boulby
  - *What is the cost of set-up and maintenance?*
  - *Can it feasibly be established in Boulby?*
  - *Scope: Are future/current experiments interested in the capability?*

  } Costing and feasibility: ECuME expertience
- Survey ongoing to assess UK interest in facility
  - Circulated to DM-UK community and in HiPhi newsletter
  - Second survey being opened to international community
- Facility would complement BUGS, especially the XIA UltraLo-1800

Ultra-Pure Copper Electroforming Capability in the UK

Rare event search experiments have been pushing the frontiers of low radioactivity for decades in the pursuit of new physics. One major development in recent years is the use of high-radiopurity electroformed copper, which suppresses typical contaminants including those from the U238/Th232 and Rn222 decay chains. More recently, underground electroforming is being undertaken to also suppress cosmogenic activation. In particular, the underground laboratories of Modane, Canfranc and Snolab are exploring electroforming. The ECuME project, funded in Canada, will establish a large-scale deep underground electroforming facility in Snolab.

This survey is to gauge the relevance for the UK rare-event search community of an underground electroforming facility at the Boulby Underground Laboratory.

https://www.surveymonkey.co.uk/r/7JLT2DW

The survey is still open, if you would like to provide input (<2 mins to complete)

Summary

- Copper has desirable qualities as detector material
- **High reduction potential** allows copper to be **purified** in electroforming process
  - **Unparalleled radiopurity** achieved through this process
    - Purity below sensitivity of world-leading assay techniques
- **Electroforming underground suppresses cosmogenic activation**
- Several underground laboratories exploring technology
- Explored by future experiments
- ECuME will be the deepest and largest electroforming capability in the world!
  - UK has unique opportunity to capitalise on this investment!