Ultra-Trace Radioassay using ICP-MS at Boulby Underground Laboratory

Science and Technology Facilities Council

Boulby Underground Laboratory

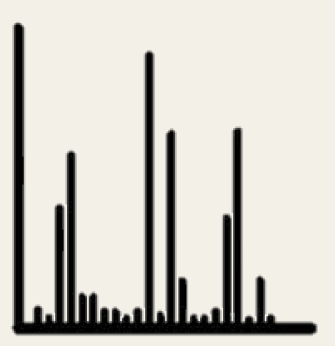
UCL

King's College London

in collaboration with University College London and King's College London

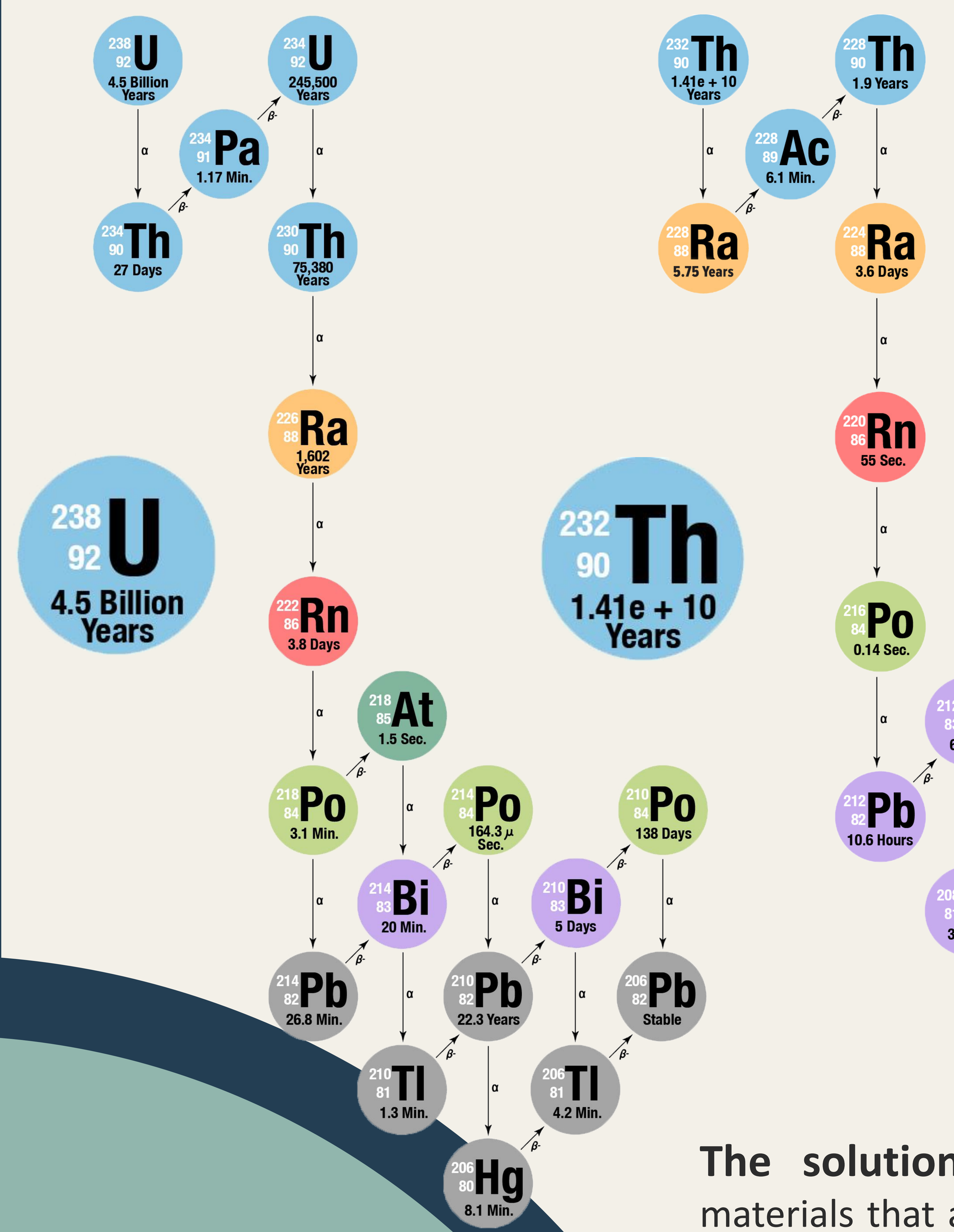
Taking Mass Spectrometry to Higher Peaks

The Inductively Coupled Plasma-Mass Spectrometer at Boulby Underground Laboratory runs in collaboration with University College London and King's College London and plays an important role in the development of high sensitivity, low detection limit particle physics. In a world where the search for rare event interactions from elusive particles is requiring more sensitive equipment, the radiopurity of the materials, electronics and surrounding environments of the particle detectors must be rigorously tested to reach their background goals.



The ICP-MS is part of the BUGS (Boulby UnderGround Screening) facility in a certified ISO 6 cleanroom which works alongside 8 germanium detectors, 2 XIA-UltraLo 1800 and a Radon Emanation system that are homed in a Class 1000 cleanroom in the underground laboratory. The ICP-MS requires an extremely clean and low particle environment, reaching consistent levels of 0 particles above 5 μm^{-3} which is imperative due to the nature of mass spectrometry; counting atoms.

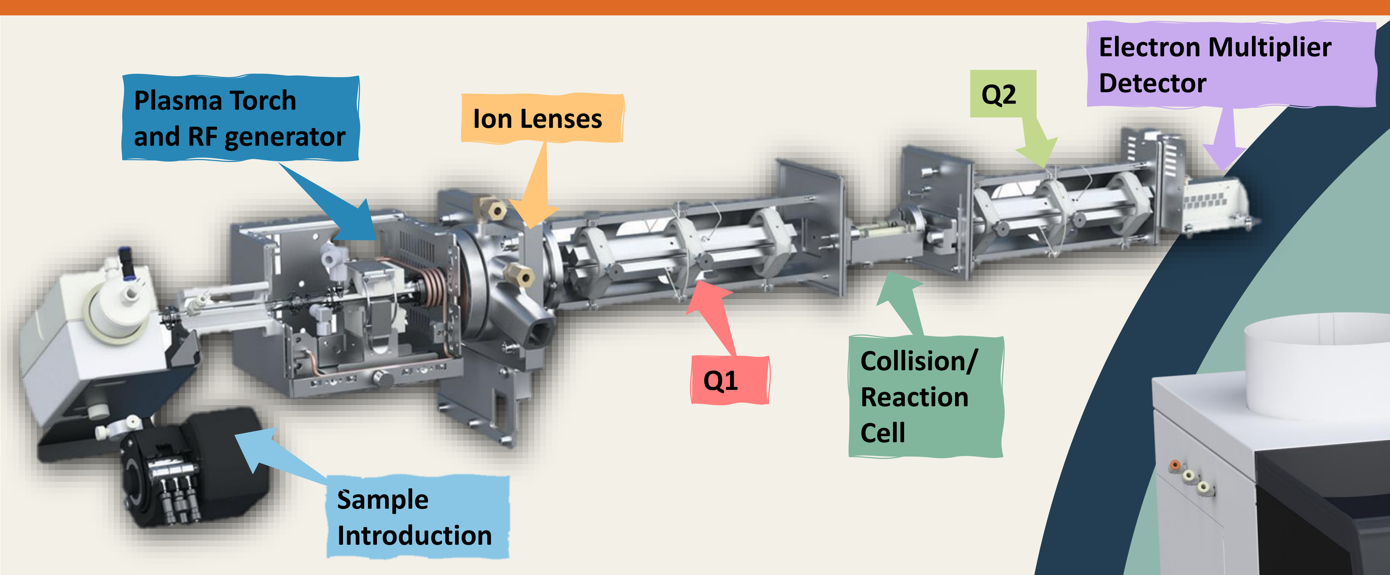
A Low Background Detectors Biggest Opp



For low background and rare event 0 detectors to work, they must be extremely radiopure, meaning that they don't have large contamination of radioactive nuclides. Naturally occurring uranium-238 (99.27% abundance) and thorium-232 (99.28%) exist everywhere in our environment such as in rocks, ores, soil, water and the air. These isotopes have very long half lives on the scale of billions of years, meaning they stick around for a long time, slowly decaying into other elements as shown in the decay chains. This becomes a problem for low background particle detectors as it creates background noise – a fog that rare event detections could be lost in. Therefore the search for dark matter, neutrino physics and neutrinoless double-beta decay experiments rely on low background techniques!

The solution? It's important to build detectors out of materials that are low in U-238 and Th-232 as to give the best chance of detecting the target particle and not a false result from a radioactive decay from contaminants.

So How Does It Work?



Sample Introduction

A sample is first digested in ultra-pure acids such as nitric, hydrochloric and hydrofluoric before being turned into an aerosol and fed into the ICP-MS where it become ionised in an argon plasma at roughly 8000 degrees.

Ion Lenses

Once ionised, the ions are carried through ion lenses which extract positive ions from the plasma beam and accelerate them into the high vacuum system.

Quadrupole 1

The ions are then beamed into the first quadrupole mass filter which only allows the target mass to charge ratio through while other masses are removed. However, this does mean interfering ions and compounds of the same mass to charge ratio can pass through too.

Collision/Reaction Cell

To remove these interferences of the same mass to charge ratio, a gas is introduced to interact with them, stopping them from passing through the next mass filter.

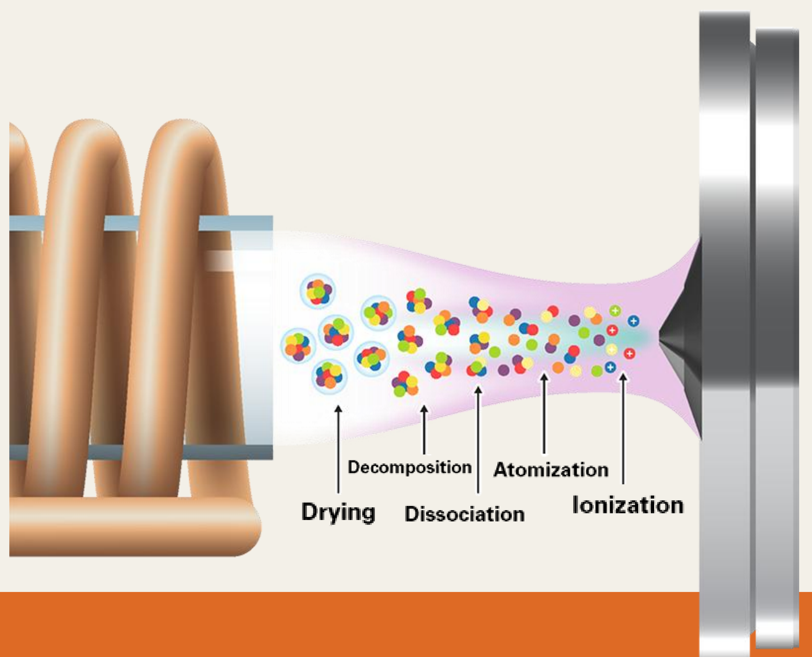
Quadrupole 2

Any remaining background ions are filtered out in the second quadrupole so only the initial target analytes are allowed through to the detector.

Detector

The U-238 and Th-232 ions pass to the electron multiplier detector for quantification, providing a counts per second output.

This is an 8000-degree argon plasma from the plasma torch!



The Menaces

222 86 Rn 3.8 Days

Radon gas emanates from materials, escaping into the surrounding environment before it decays, plating onto surfaces and eventually decaying into lead

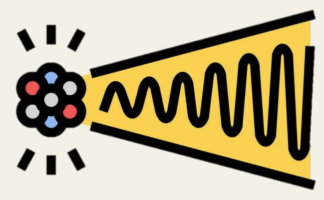
210 84 Po 138 Days

214 84 Po 164.3 µ Sec.

Polonium isotopes are alpha emitters, creating unwanted signals

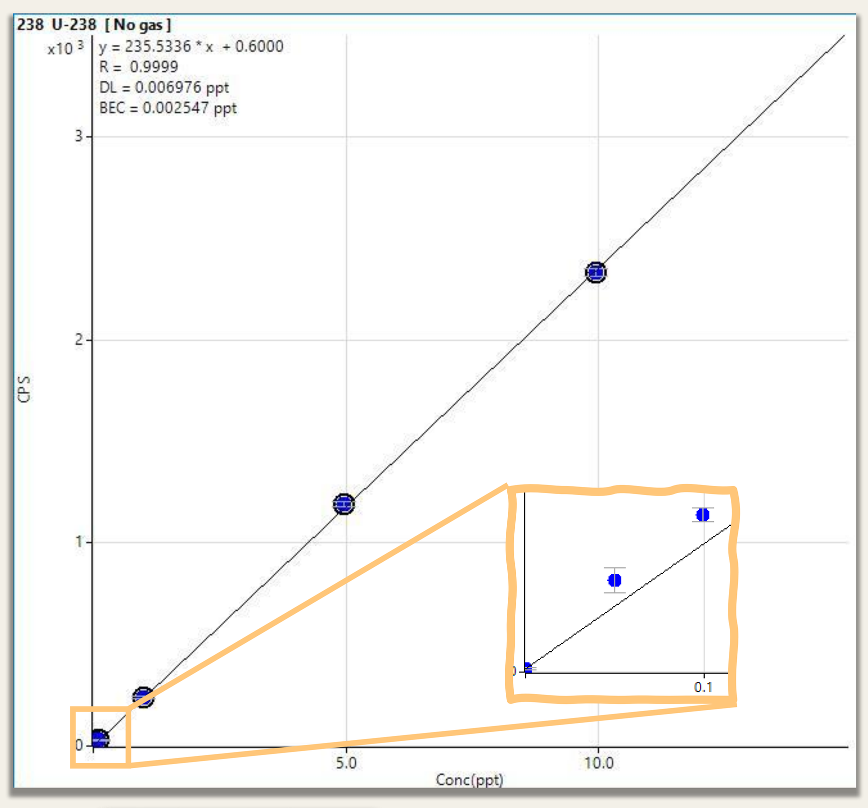
210 82 Pb 22.3 Years

Down the U-238 decay chain, Lead-210 hangs around for 22.3 years before decaying, causing many years of background noise in a detector!



Gamma radiation emitted down the decay chains is bad news for neutrinoless double-beta decay experiments!

Calibrations, Blanks and Samples



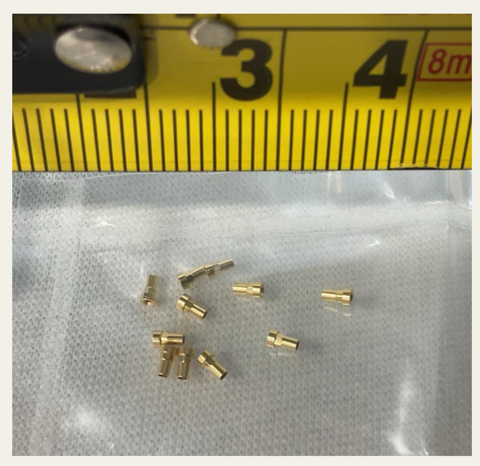
Blank solutions are analyte free versions of the same acids and concentrations used in the sample digestion. This establishes a zero point on the calibration curve, monitors contamination and corrects for background interferences.

Calibration standards correlate counts per second to known concentrations. Calibration standards are made by adding known concentrations of U-238 and Th-232 to a blank solution, analysing each standard and generating this graph. The CPS reading from analysing a sample is then converted to a concentration using this graph.

Conc.	Calc Conc.	CPS
0.000	0.000	0.60
0.050	0.071	17.36
0.100	0.123	29.54
0.996	1.015	239.73
4.970	5.062	1192.89
9.950	9.902	2332.79

This is the blank. Despite having no U or Th added, there are still counts!

These are the calibration standards shown on the graph, ranging from 10 – 0.05ppt



We only need a very small sample up to around 200mg to get results. We can measure the uranium and thorium concentrations in these individual PMT pins! However, it is a destructive technique so the sample will be lost after analysis.

What's Next?

40 19 K Potassium-40 1.248 Billion Years

- Expand the analytes that we trace, moving beyond U-238 and Th-232 and looking into potassium-40 which will prove challenging due to the large number of interferences that can occur.
- Push for better detection limits and increased sensitivity by commissioning a prepFAST-MC ion exchange chromatography system to remove the matrix (acid and other elements the U and Th is dissolved in) and leaving behind the target analyte.
- Reach beyond the scope of often required material radiopurity thresholds of 1 - 0.01 ppt and achieve ultra-trace sensitivities of 10 ppq.

TL;DR

- **Ultra-trace radiopurity matters:** U-238 and Th-232 creates persistent background signals potentially obscuring rare-event measurements
- **ICP-MS enables ultra-trace sensitivities:** We can quantify extremely low concentrations of U/Th, supporting material selection for low background experiments
- **Cleanliness is critical:** Ultra-clean environment and equipment, high purity acids and rigorous control of blanks is essential
- **Supporting major collaborations:** We support collaborations such as XLZD and LEGEND!

References: www.nachi.org/gallery/radon/thoron-decay-chain | www.agilent.com | www.spectro.com/inductively-coupled-plasma-mass-spectrometry-icp-ms-principle

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