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Ultra-low background mass spectrometry for rare-event searches

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ABSTRACT

Inductively Coupled Plasma Mass Spectrometry (ICP-MS) allows for rapid, high-sensitivity determination of trace impurities, notably the primordial radioisotopes ^{238}U and ^{232}Th, in candidate materials for low-background rare-event search experiments. We describe the setup and characterisation of a dedicated low-background screening facility at University College London where we operate an Agilent 7900 ICP-MS. The impact of reagent and carrier gas purity is evaluated and we show that twice-distilled ROMIL-SpA^{TM}-grade nitric acid and zero-grade Ar gas delivers similar sensitivity to ROMIL-UpA^{TM}-grade acid and research-grade gas. A straightforward procedure for sample digestion and analysis of materials with U/Th concentrations down to 10 ppt g/g is presented. This includes the use of ^{233}U and ^{230}Th spikes to correct for signal loss from a range of sources and verification of ^{238}U and ^{232}Th recovery through digestion and analysis of a certified reference material with a complex sample matrix. Finally, we demonstrate assays and present results from two sample preparation and assay methods: a high-sensitivity measurement of ultra-pure Ti using open digestion techniques, and a closed vessel microwave digestion of a nickel–chromium-alloy using a multi-acid mixture.

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1. Introduction

Rare event search experiments such as those seeking interactions from galactic dark matter scattering or evidence of neutrino-less double beta decay ($0\nu\beta\beta$) require construction from intrinsically radio-pure materials to limit radiogenic backgrounds. Experiments must conduct radio-assay campaigns to select materials and to accurately characterise residual radioactivity for the experiment's background model against which any observed excess will be evaluated and statistical confidence ascribed. The radioactive isotopes ^{238}U and ^{232}Th and their decay-chain progeny are of particular concern, contributing the bulk of the γ-ray, α-particle, and, via spontaneous fission and (α, n) reactions, neutron-induced backgrounds.

Comprehensive radio-assay campaigns now increasingly deploy multiple techniques to build a high-precision background model, and to meet sample throughput requirements during the experiment's construction phase. Mass spectrometry techniques such as ICP-MS can be used to directly measure trace quantities of ^{238}U and ^{232}Th, delivering assays from small samples that are digested and screened with turnaround times on the order of 1–2 days. ICP-MS can achieve part-per-trillion (ppt) g/g level sensitivity to ^{238}U and ^{232}Th when sample preparation protocols with stringent cleanliness constraints are used to limit contamination [1–3]. Gamma-spectroscopy is used to assay larger samples, including finished components, over several weeks to determine the activities of the decay chain daughters [4]. Other techniques such as Glow-Discharge Mass Spectrometry (GD-MS) and Neutron Activation Analysis (NAA) may also be used for some materials not well-suited to either ICP-MS or gamma-spectroscopy.

Here we present results from a new ICP-MS facility at University College London (UCL) constructed to perform radio-assays in support of rare-event searches, such as the 'Generation-2' LUX-ZEPLIN (LZ) dark matter Experiment [5]. In Section 2 we give an overview of the UCL ICP-MS facility and investigate the impact of reagent and Ar gas purity on achievable sensitivity. Then, in Section 3, we describe a straightforward procedure for routine and fast-turnaround of materials with U/Th concentrations down to 10 ppt g/g. We present detection limits for different vessel cleaning protocols and describe the use of ^{230}Th and ^{233}U spikes for signal loss correction. A certified reference material representing a realistic sample matrix is processed to verify overall U/Th signal recovery. Finally, in Section 4 we present results from an open digestion of ultra-pure Ti and a closed vessel digestion of a nickel–chromium-alloy requiring multi-acid digestion.

2. Laboratory overview

The UCL ICP-MS facility was installed and commissioned in a newly constructed ISO Class 6 cleanroom in late 2015. The primary instrument is an Agilent 7900 ICP-MS with an instrument sensitivity to U and Th

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down to 10^{-15}–10^{-14} g/g (1–10 parts-per-quadrillion (ppq) g/g). The ICP-MS is fitted with an integrated auto-sampler for high-speed discrete sample uptake with a low-flow, Peltier-cooled sample introduction system. High-purity Ar (N5.5 or N5.0), He and H_2 (N6.0) are introduced to the system as carrier, collision and reaction gases, respectively. The system octopole can provide species discrimination through interference removal by running in either the He collision or H_2 reaction gas mode. The system is fitted with an inert sample introduction kit that includes a PFA nebuliser, spray chamber and torch connector, platinum sampling and skimmer cones and a plasma torch with quartz outer body and sapphire injector. This allows up to 20% (v/v)[1] acid concentration in the sample introduced to the ICP-MS, including hydrofluoric acid (HF).

The limiting factor in realising reproducible high throughput ppt sensitivity is clean, standardised sample preparation requiring dedicated digestion apparatus and well developed procedures, including the use of ultra-pure acids to avoid contamination of samples. The facility has the relevant infrastructure to provide this, including: sample preparation in separate ISO Class 5 laminar flow unit (LFU), sample digestion with the Milestone ETHOS-UP closed vessel microwave digestion system with SK-15 high pressure rotor, Milestone sub-boiling point acid distillation (subCLEAN) and reflux cleaning (traceCLEAN) systems and an ELGA PURELAB flex ultra-pure de-ionised (DI) water supply (18.2 MΩ. cm, <5 parts-per-billion (ppb) total organic carbon). Finally, a Pyro-260 microwave ashing system allows for the digestion of materials such as PTFE that are resistant to most acids, including HF.

For most assays we follow material-by-material digestion routines available for the ETHOS-UP and SK-15 high pressure rotor. These specify the acid combinations and microwave heating profiles needed to achieve complete digestion. Where these do not exist, custom routines for non-standard materials have been developed based on existing routines and in partnership with Milestone and U.K. supplier Analytix Ltd. Typically nitric acid (HNO_3) is used to digest organic material, hydrofluoric acid (HF) for decomposing silicates and metals such as Ti that are resistant to oxidising acids, and hydrochloric acid (HCl) for Fe-based alloys. When fitted with the SK-15 high pressure rotor the ETHOS-UP can simultaneously digest up to 15 samples (with the same chemistry) each in their own 100 mL TFM vessels. TFM is a chemically modified form of PTFE with excellent properties for trace analysis: high chemical resistance, low-trace metal impurities, high melting point and an extremely smooth surface for decontamination and cleaning.

2.1. Reagent purity

The purity of acids and other reagents used for digestion, vessel-cleaning and preparation of calibration standards is critical for the limit of detection achievable when assaying a material. For standard cleaning of vessels and digestion of samples with ≫ ppb U/Th concentrations we use ROMIL-SpA™ Super Purity Acids (SpA). These are cost effective acids produced using sub-boiling point distillation and are typically certified to <100 ppt ^{238}U and ^{232}Th per gram of reagent. For lower detection limits and sub-ppb materials we use ROMIL-UpA™ acids and reagents which are produced through multiple re-distillation and are typically certified to <0.1 ppt ^{238}U and ^{232}Th.

Table 1 compares measured background equivalent concentrations (BEC) for nitric acid process blanks made with ROMIL-SpA and ROMIL-UpA acids as well as for ROMIL-SpA nitric acid that was subsequently twice-distilled using the in-house subCLEAN distillation system. Inferred concentrations for undiluted HNO_3 are consistent with the manufacturer's stated typical concentrations: <100 ppt U/Th for SpA and <0.1 ppt U/Th for UpA. We find that twice-distilled SpA is consistent with the UpA background concentration for ^{232}Th and approaches that for ^{238}U. Based on these results we now use twice-distilled SpA-grade nitric for most digestions unless ultimate detection limits are required. In addition, twice-distilled SpA can be used in cleaning procedures requiring larger quantities of reagents where UpA-grade acid is prohibitively costly.

[1] Unless stated otherwise v/v indicates a dilution by volume of reagent at standard concentration, in this case 69% HNO_3, with de-ionised water.

Table 1
^{232}Th and ^{238}U BECs for different grades of ROMIL nitric acid diluted to ~20% (v/v) with DI water. Solutions were prepared in 50 mL PP (polypropylene) vessels pre-leached in SpA nitric acid for 24 h. Equivalent concentrations for concentrated 69% nitric acid are shown. These represent upper limits on the acid purity as contributions from other sources, such as leaching from vessels, are not subtracted.

	ppt g/g ~20% (v/v) diluted	
	^{232}Th	^{238}U
ROMIL-SpA	1.730 ± 0.160	1.85 ± 0.06
ROMIL-UpA	0.035 ± 0.005	0.03 ± 0.01
twice-distilled-SpA	0.030 ± 0.004	0.05 ± 0.01
	Eqv. ppt g/g 69% HNO_3	
ROMIL-SpA	6.61 ± 0.60	7.06 ± 0.25
ROMIL-UpA	0.12 ± 0.02	0.10 ± 0.03
twice-distilled-SpA	0.11 ± 0.02	0.19 ± 0.04

Table 2
HNO_3 blank rates for different Ar gas supplies using 5% (v/v) HNO_3 (ROMIL UpA). For the second two measurements all labware (PP vials and digiTubes) were leached in 10% ROMIL SpA HNO_3 for 24 h prior to use. For each run the instrument response based on external calibrations were ~300 CPS/ppt.

ppq response 5% HNO_3 blank		
	^{232}Th	^{238}U
N5.5 Ar	13.5 ± 1.4	11.8 ± 1.8
N5.5 Ar (leached)	7.1 ± 0.9	5.9 ± 1.9
N5.0 Ar (leached)	9.1 ± 2.2	9.2 ± 2.3

2.2. Ar gas supply

In analysis mode the ICP-MS consumes up to 20 litres per minute of argon gas, one 50 L 200 bar cylinder for every 8 h of running.[2] Given the importance of Ar gas purity for ultra-trace analysis and the potential costs associated with the high-turnaround we have compared the performance of the ICP-MS with two different grades of bottled gas: N5.0/zero-grade (min. 99.999%) and N5.5/research-grade (min. 99.9995%), where the cost of research-grade is significantly higher, between 5 and 10 times that of zero-grade from our supplier.

Table 2 shows the ^{232}Th and ^{238}U BECs for a 5% (v/v) HNO_3 acid blank for both zero- and research-grade argon. Two sets of acid blanks were prepared, a first using unconditioned labware (virgin PP DigiTubes and 1 L Nalgene containers) that was simply rinsed with DI water prior to use and a second set where all labware was first leached in 10% (v/v) SpA HNO_3 for 24 h prior to use. Switching to blanks prepared using pre-conditioned labware halved the BECs down to 7.1 ppq ^{232}Th and 5.9 ppq ^{238}U when using research-grade argon. Zero-grade argon shows a few ppq increase with respect to research-grade although results are almost within measurement errors.

Switching to zero-grade did not cause deterioration in the CPS/ppt instrument response or the level of oxides and doubly charged species during tuning; in this case ($^{156}CeO^+/^{140}Ce^{++}$) and ($^{70}Ce^{++}/^{140}Ce^+$). Given this and the sub-dominant blank rate increase we conclude zero-grade argon is sufficient for most routine samples where ^{232}Th and ^{238}U are the analytes of interest.

3. Procedures and analysis

With a focus on high-throughput and fast-turnaround of time-critical assays, straightforward cleaning, sample preparation and analysis procedures have been developed for materials with U/Th concentrations in the 10 ppt to 1 ppb range: Samples are microwave-digested in pre-cleaned TFM vessels using ultra-high purity acids. They are then diluted, without further chemical treatment, into disposable 50 mL polypropylene (PP) vessels ready for ICP-MS analysis. Fractional recoveries of

[2] A liquid argon supply was considered but was less cost effective due to wastage during periods of downtime.

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