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Reduction of radioactive backgrounds in electroformed copper for ultra-sensitive radiation detectors

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ABSTRACT

Ultra-pure construction materials are required for the next generation of neutrino physics, dark matter and environmental science applications. These materials are also important for use in high-purity germanium spectrometers used in screening materials for radiopurity. The next-generation science applications require materials with radiopurity levels at or below 1 μBq/kg ^{232}Th and ^{238}U. Yet radiometric analysis lacks sensitivity below ~10 μBq/kg for the U and Th decay chains. This limits both the selection of clean materials and the validation of purification processes. Copper is an important high-purity material for low-background experiments due to the ease with which it can be purified by electrochemical methods. Electroplating for purification into near-final shapes, known as electroforming, is one such method. Continued refinement of the copper electroforming process is underway, for the first time guided by an ICP-MS based assay method that can measure ^{232}Th and ^{238}U near the desired purity levels. An assay of electroformed copper at a μBq/kg level has been achieved and is described. The implications of electroformed copper at or better than this purity on next-generation low-background experiments are discussed.

1. Introduction

The success of modern experiments to detect neutrino recoils [1], double-beta decay [2,3], dark matter [4], radiopurity assay in support of these, and studies of terrestrial and extra-terrestrial constituents for many purposes Laubenstein et al., Plastino and Kaihola, Hult et al. and Arnold et al. [5–8] all depend on materials selected for purity, or purified through a chemical process. Some materials of the desired purity can be found, such as those recovered from sources of ancient natural gas uncontaminated by cosmic-ray spallation or debris from anthropogenic sources such as nuclear weapons explosions and nuclear accidents. Other materials must undergo extensive purification before they can be included in detectors. Thus, a focused effort to create ultra-pure material using a chemical purification process has intensified to respond to the need for ever-purer construction materials.

1.1. The challenge of greater purity

Sources of obscuring signals, e.g., background, in radiometric measurements can be classified into three areas:

a. Signals arising from cosmic rays.
b. Signals arising from radioactivity in the detector and shield system.
c. Signals created by uninteresting radioactivity within the sample being measured.

Moving a detector system underground is an excellent strategy to lower the overall background if cosmic-ray-associated background is a large contributor. However, Laubenstein et al. [5] presented an intriguing analysis of the full-spectrum count rate of several detectors in which below ~500 m water equivalent (mwe) increasing depth did not result in a decreasing trend in background count rate. One reasonable conclusion is that materials in the detector and shield were the dominant source of background below that depth. Counting rates from additional detector systems developed by Pacific Northwest National Laboratory (PNNL) and collaborators are shown in Fig. 1 with similarly integrated count

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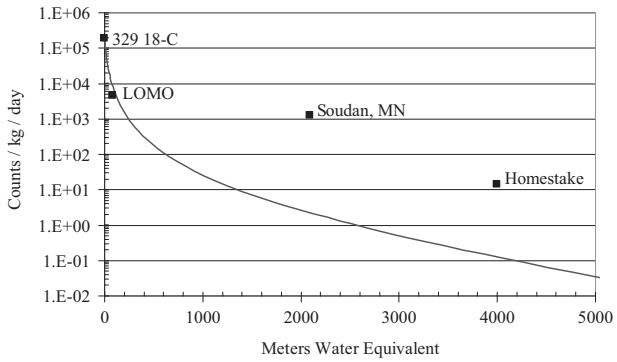

Fig. 1. Comparative integrated counting rates similar to [5] from 40 to 2700 keV for several PNNL systems: located at PNNL (329 18-C); under Lower Monumental Dam (LOMO, 75 mwe) near Richland, WA; in the Soudan mine near Soudan, MN; and at the 4850 foot level of the Homestake mine, near Lead, SD. An arbitrarily scaled muon fluence curve is shown for reference, computed as in Heisinger et al. [24].

rates. This generally strengthens the conclusion that the majority of the background results from materials contamination, even though these detector systems have little in common in their production process with those discussed in Laubenstein et al. [5].

One might conclude that overall detector purity needs to be from 100 to 1000 times better to take full advantage of deep underground science laboratories, i.e., spaces deeper than 500 mwe. In reality, materials contamination is likely spread unevenly between a variety of components such as structural materials, external gamma ray shielding, surface coatings, electronics, and other components. In addition to materials contamination, surface contamination also has the potential to be a major contributor and can be impacted by handling, reagents, aerosols, or by exposure to ambient radon and its decay products. Because of this, clean rooms, radon-scrubbed air, reduced exposure time, highly purified reagents, and other surface mitigation preparations are important [9,10]. For contaminants that include alpha decay, this can strongly influence the selection of detector technology to minimize surface alpha sensitivity, e.g., p-type vs. n-type high purity germanium detectors (HPGe).

For this work, the focus is the reduction of impurities in copper. Copper can be the most massive component in many ultra-low-background cryostats including those produced by PNNL for experiments such as the Majorana Demonstrator [11]. However, copper radioactivity reduction should be considered in the context of a complete background budget, and a comparison will be made between copper contributions and other materials at published radioactivity levels.

2. Methods for the production of pure copper

Unfortunately, most desirable materials for detector parts and cryostat construction suffer from one or more modes of contamination. Copper suffers activation due to reactions with cosmic-ray-produced secondary neutrons (e.g., $^{63}Cu(n,\alpha)^{60}Co$) as well as contamination with U and Th during the production and handling processes. Perhaps most damaging is the creation of ^{60}Co ($T_{½}=5.2$ year, $Q_{\beta}=2.5$ MeV) in copper. Although some excellent sources of commercial copper can be found, the effects of cosmic-ray exposure are long-lived and preventing activation can be complex [12]. Thus, earlier basic copper purification efforts for cryostat and shielding purposes were carried out [13].

2.1. The electroforming method

The electroplating process is shown notionally in Fig. 2, similar to that reported previously [13]. Two processes ("Reeves" and "Hoppe") have been developed in parallel with a common assay method. The voltage and current plating waveform, temperature, conductivity, and other factors of the bath are monitored and controlled [14]. However, monitoring progress in the rejection of trace impurities can be quite difficult at the levels of interest, where levels of ^{232}Th or ^{238}U of 1 μBq/kg copper (or 0.25 pg ^{232}Th/g and 0.08 pg ^{238}U/kg Cu) or better are required. Direct radiological measurements employing ~10 kg of copper measured for ~100 days on the world's lowest background gamma-ray assay detectors have resulted in limits on the ^{232}Th chain from less than 9 μBq/kg to less than 28 μBq/kg [15–18].

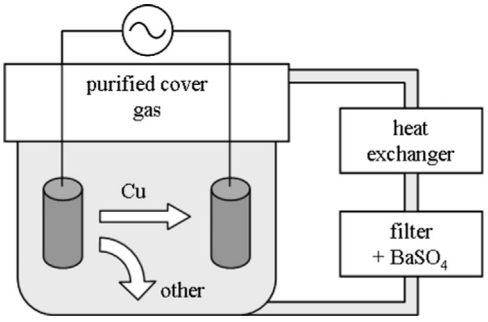

Fig. 2. Notional diagram of the electroplating process shows the movement of copper ions across a H_2SO_4 /$CuSO_4$ bath under the influence of a low-voltage. A filtration cartridge for particulates is also seeded with $BaSO_4$ for the purpose of scavenging Ra from the solution.

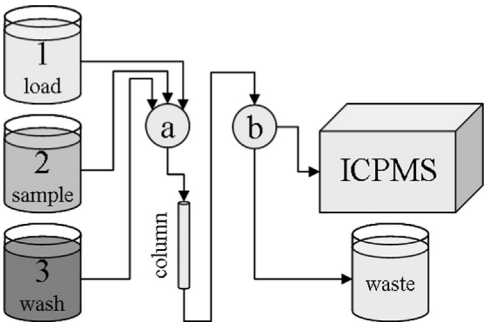

Fig. 3. The ICP-MS procedure uses a conditioning (load) solution (1), a sample solution at the same pH (2), and then an eluting solution (3) into valve (a) After the sample has passed through the column, the wash solution strips the Th from the exchange resin and through valve (b) to produce a concentrated pulse into the ICP-MS. A mass 229 tracer indicates the timing of the mass 232 pulse.

2.2. Using ICP-MS to measure thorium and uranium

To obtain assay sensitivity well below 10 μBq/kg in a reasonable time frame, an inductively coupled-plasma/mass spectrometry (ICP-MS) approach was explored. At first glance, this may seem straightforward, since instrumental sensitivity can be as much as a thousand times better than the goal. Unfortunately, reagents, containers, tubing, dust, and ion exchange resins can contribute to a ubiquitous background of Th and U at obscuring levels. For example, some lab ware was found to reliably yield a small quantity of Th and U at each leaching with ultra-pure reagent, even after dozens of leaching preparation steps. Many of the materials in the process, notably plastic tubing inside the system, can function like exchange resins, retaining and releasing Th at varying pH. In addition, since the column material is designed to bind Th, it is always possible that some residual Th will be released in the elution step [19]. The notional sample preparation and ICP-MS scheme is shown in Fig. 3.

To estimate the sensitivity of the assay system, process blanks were frequently measured during the analysis of samples. The samples included both starting anode and the electroformed