Copper electroplating for background suppression in the NEWS-G experiment

L. Balogh\textsuperscript{a}, C. Beaufort\textsuperscript{b}, R. Bunker\textsuperscript{c}, A. Brossard\textsuperscript{a}, J.-F. Caron\textsuperscript{a}, M. Chapellier\textsuperscript{a}, E.C. Corcoran\textsuperscript{d}, J.-M. Coquillat\textsuperscript{a}, S. Crawford\textsuperscript{a}, A. Dastgheibi Fard\textsuperscript{b}, Y. Deng\textsuperscript{c}, K. Dering\textsuperscript{a}, D. Dunford\textsuperscript{e}, I. Giomataris\textsuperscript{f}, P. Gorel\textsuperscript{g,h,i}, M. Gros\textsuperscript{j}, P. Gros\textsuperscript{a}, G. Gerbier\textsuperscript{a}, G. Giroux\textsuperscript{g}, O. Guillaudin\textsuperscript{b}, E.W. Hoppe\textsuperscript{c}, I. Katsioulas\textsuperscript{j}, F. Kelly\textsuperscript{d}, P. Knights\textsuperscript{f,j}, L. Kwon\textsuperscript{d}, S. Langrock\textsuperscript{h}, P. Lautridou\textsuperscript{k}, J.-P. Mols\textsuperscript{f}, R. D. Martin\textsuperscript{a}, J.-F. Muraz\textsuperscript{b}, X.-F. Navick\textsuperscript{f}, T. Neep\textsuperscript{j}, K. Nikolopoulos\textsuperscript{j}, P. O’Brien\textsuperscript{e}, R. Owen\textsuperscript{j}, M.-C. Piro\textsuperscript{a}, G. Savvidis\textsuperscript{a}, I. Savvidis\textsuperscript{l}, F. Vazquez de Sola Fernandez\textsuperscript{a}, M. Vidal\textsuperscript{a}, R. Ward\textsuperscript{j}, M. Zampaolo\textsuperscript{b}

(NEWS-G Collaboration)

Abstract

New Experiments with Spheres-Gas (NEWS-G) is a dark matter direct detection experiment that will operate at SNOLAB (Canada). Similar to other rare-event searches, the materials used in the detector construction are subject to stringent radiopurity requirements. The detector features a 140-cm diameter proportional counter comprised of two hemispheres made from commercially sourced 99.99% pure copper. Such copper is widely used in rare-event searches because it is readily available, there are no long-lived Cu radioisotopes, and levels of non-Cu radiocontaminants are generally low. However, measurements performed with a dedicated $^{210}$Po alpha counting method using an XIA detector confirmed a problematic concentration of $^{210}$Pb in bulk of the copper. To shield the proportional counter’s active volume, a low-background electroforming method was adapted to the hemispherical shape to grow a 500-µm thick layer of ultra-radiopure copper to the detector’s inner surface. In this paper the process is described, which was prototyped at Pacific Northwest National Laboratory (PNNL), USA, and then conducted at full scale in the Laboratoire Souterrain de Modane in France. The radiopurity of the electroplated copper was assessed through inductively coupled plasma mass spectrometry (ICP-MS). Measurements of samples from the first (second) hemisphere give 68% confidence upper limits of $<0.58$ µBq/kg ($<0.24$ µBq/kg) and $<0.26$ µBq/kg ($<0.11$ µBq/kg) on the $^{232}$Th and $^{238}$U contamination levels, respectively. These results are comparable to previously reported measurements of electroformed copper produced for other rare-event searches, which were also found to have low concentration of $^{210}$Pb consistent with the background goals of the NEWS-G experiment.

Keywords: Dark matter, Direct detection, Rare Event, Electroforming, Copper, Low background
Figure 1: Schematic diagram of the NEWS-G detector and shielding. The $\phi 140$ cm spherical proportional counter is shown at the centre, surrounded by 3 cm of archaeological lead, followed by 22 cm of low radioactivity lead in a stainless steel skin. The outer-most part of the shielding comprises 40 cm of high-density polyethylene (HDPE).

1. Introduction

Direct searches for dark matter (DM) and neutrinoless double-beta decay [1, 2, 3, 4] have strict requirements on the experimental background to achieve their targeted sensitivities. While such experiments are generally carried out in underground laboratories and in specifically designed shielding to suppress backgrounds from external sources, one of the main remaining sources arises from radioactive decays in the detector’s construction materials, including the gaseous target. The effort to procure materials with the lowest possible radioactivity has driven significant improvements in the techniques and facilities used to assay and prepare radiopure materials [5, 6, 7, 8].

A common choice for a high-purity material is commercially sourced copper [9, 10, 11], because it is readily available and there are no long-lived Cu radioisotopes—with a half-life of 61.8 hours [12]. $^{67}$Cu is the longest-lived. For this reason, the NEWS-G collaboration [13] chose 4N (99.99% pure) copper to construct a $\phi 140$ cm spherical proportional counter [14], which will be housed in the compact shielding shown in Fig. 1 to perform a direct DM search at SNOLAB, Canada. Even without long-lived Cu radioisotopes, a copper sample will have some (non-Cu) radiogenic contamination resulting from cosmogenic activation and industrial production processes. For example, cosmic-ray neutrons interacting with copper through the $$(n, \alpha)$$ reaction can produce $^{60}$Co. The half-life of the produced $^{60}$Co is approximately 5.3 years, making it a long-lived background relative to the typical time scale of direct DM detection experiments. At the surface of the Earth, the added activity due to $^{60}$Co is approximately 0.4 $\mu$Bq/kg/day [15]. Other cosmogenic contaminants with shorter half-lives are also produced, e.g. $^{59}$Fe. These contributions can be suppressed by minimising the copper’s exposure to cosmic rays. Other radiocontaminants primarily originate from the $^{238}$U and $^{232}$Th decay chains. The $^{238}$U decay chain is shown in Fig. 2. This contamination is inherent to the raw material and a result of the manufacturing and handling processes. An established technique is to directly measure the uranium and thorium levels with inductively coupled plasma mass spectrometry (ICP-MS), which has been demonstrated to have sensitivity better than 30 fg/g to these contaminants [6, 16, 17]. The progeny activities can also be inferred and used to estimate background contributions to experiments, under the assumption of secular equilibrium.

However, $^{222}$Rn, which is part of the $^{238}$U de-
The estimation of $^{210}\text{Po}$ is expected to be in the range of 17–40 mBq. An approach to suppress the background expected is by directly measuring the $^{210}\text{Po}$ decays, using a high-sensitivity XIA UltraLo-1800 spectrometer, which has a sensitivity of 0.0001 $\alpha$/cm$^2$/hour. The XMASS collaboration has established a method to estimate very low $^{210}\text{Po}$ contamination in copper bulk, having demonstrated the ability to distinguishing the contamination in bulk from that on the surface. For oxygen-free copper (at least 99.96% pure by weight) the $^{210}\text{Po}$ contamination is estimated to be in the range of 17–40 mBq/kg. The estimation of $^{210}\text{Po}$ contamination in the 4N copper procured by the NEWS-G collaboration is discussed in Section 2.

This level of $^{210}\text{Po}$ in the bulk of the NEWS-G detector’s copper sphere would dominate the expected background and define the experimental sensitivity. An approach to suppress the background from $^{210}\text{Po}$ contamination is to grow a layer of ultra-radiopure copper onto the inner surface of the detector sphere. This layer acts as an internal shield to suppress backgrounds, e.g., from $\beta$-decays of $^{210}\text{Po}$ and accompanying X-rays and Auger electrons, and its progeny $^{210}\text{Bi}$, originating from the bulk of the commercially sourced 4N copper. It is estimated by means of a Geant4 simulation that a 500 µm-thick layer of ultra-radiopure copper will suppress the background in the energy region of interest by approximately 62% to 1.04 $\pm$ 0.38 counts/keV/kg/day.

A method to deposit ultra-radiopure copper is potentiostatic electroforming. This method takes advantage of electrochemical properties to produce copper with reduced impurities. The process is described in Section 3. This method was previously used to produce a variety of detector components, including those requiring extreme radiopurity such as for the Majorana Demonstration.

$^{210}\text{Po}$ has a half-life of approximately 138 days, which is significantly shorter than the approximately 22 years of the progenitor $^{210}\text{Pb}$. As a result, the activities of $^{210}\text{Po}$ and $^{210}\text{Pb}$ may be different due to different contamination amounts at the production phase. Therefore, the activities of the two isotopes may be out of secular equilibrium; however, the $^{210}\text{Po}$ activity in a sample will evolve over time.

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3 Japanese Industrial Standard, JIS:C1020

4 For $^{238}\text{U}$, 1 µBq/kg $\approx$ 0.081 pg/g. For $^{232}\text{Th}$, 1 µBq/kg $\approx$ 0.244 pg/g.
until it matches that of $^{210}\text{Pb}$. Therefore, multiple measurements of the $^{210}\text{Po}$ activity over time are required to accurately infer the activity of $^{210}\text{Pb}$ in the copper. Four measurements of the $\alpha$ particles from the sample were made over the course of approximately one year, each lasting between 12 and 23 days. Table 1 shows the results of the four measurements.

Table 1: Measurements of the $\alpha$ particles in a 2.5 MeV to 4.8 MeV energy window originating from $^{210}\text{Po}$ decays in a C10100 copper sample.

| Date               | Measurement [10$^{-4}$ $\alpha$/cm$^2$/hour] |
|--------------------|-----------------------------------------------|
| Jul. 2 - 25, 2018  | 2.3 ± 0.4                                     |
| Oct. 5 - 17, 2018  | 2.2 ± 0.4                                     |
| Dec. 28, 2018 - Jan. 9, 2019 | 1.4 ± 0.3                               |
| Apr. 19 - May 7, 2019 | 1.4 ± 0.3                                 |

A joint likelihood fit of all measurements was performed and is shown in Figure 3 along with the measurements. From this fit, it was estimated that the $^{210}\text{Pb}$ activity in the sample is $29^{+8+9}_{-8-3}$ mBq kg$^{-1}$, where the statistical and systematic uncertainties are given separately. This is consistent with other copper samples with similar purity [20].

3. Electroplating

Electroplating is carried out through the use of an electrolytic cell, which consists of an anode and a cathode separated by an electrolyte, as illustrated in Fig. 4. A current is used to supply electrons to the cathode where an ion undergoes a reduction reaction (gain of electrons) to form an atom deposited on the cathode in reduction reactions.

$$A^{(y+z)+} + ze^- \rightleftharpoons A^y^+,$$

where $A$ is the molecular species, $y$ is its ionic charge and $z$ is the number of electrons required for the reduction reaction (reading left-to-right) or the number of electrons released in the oxidation reaction (reading right-to-left). Reading this equation in one direction gives the “half-cell reaction”, where the anode and cathode half-cell reactions are not necessarily the same; e.g., in the case where one species is oxidized at the cathode but a different species is reduced at the anode.

A current $I$ flows through the circuit and electrolyte. As the reduction reactions require electrons, the rate of electroplating is proportional to the total supplied charge: $Q(t) = \int I dt$. The number of moles $n$ of ions reduced in time $t$ is given by

$$n(t) = \frac{Q(t)}{zF},$$

where $F = eN_A$ is the Faraday constant, and $e$ and $N_A$ are the elementary charge and the Avogadro constant, respectively. The resulting deposited mass as a function of time is

$$M(t) = m_r n(t),$$
where \( m_r \) is the molecular mass of the deposited species. When the current is reversed the process is called electropolishing, which is a technique used to remove material from a surface.

There will be several species of ions in the electrolyte available to electroplate to the cathode. The tendency of an ion species to be reduced is quantified by the reduction potential \( E^0 \). Examples are shown in Table 2 for copper and radioisotope contaminants. A greater value of \( E^0 \) indicates a species that is more easily reduced. Each half-cell reaction will have its own reduction potential. The standard cell potential \( E_{\text{cell}}^0 \) of the electrolytic cell is defined as the difference between the reduction potentials of the half-cell reactions at the anode \( E_A^0 \) and cathode \( E_C^0 \):

\[
E_{\text{cell}}^0 = E_C^0 - E_A^0. \tag{4}
\]

For \( E_{\text{cell}}^0 < 0 \), additional energy will be required for the reaction to proceed \[25\]. For \( E_{\text{cell}}^0 \geq 0 \), the reaction is spontaneous (or in chemical equilibrium in the case of equality). For a given species being oxidized at the anode, the reaction will only proceed when the cathode half-cell reaction has a higher reduction potential. In the case of a copper anode being oxidized, only ion species in the electrolyte with a reduction potential greater than that of copper will reduce. The relatively high reduction potential of copper compared to many radioisotopes means that it is purified during electroplating. However, other factors, such as mass transport of contaminant ions, can cause species with lower reduction potentials to be deposited with the copper in small amounts \[23\].

### Table 2: Reduction potential for copper and possible radiocontaminants.

| Reductants  | Oxidants  | \( E^0 \) (V) |
|-------------|-----------|---------------|
| \( \text{Cu}^{2+} + 2e^- \) | \( \text{Cu} \) | +0.34 \[26\] |
| \( \text{Pb}^{2+} + 2e^- \) | \( \text{Pb} \) | −0.13 \[27\] |
| \( \text{U}^{4+} + 3e^- \) | \( \text{U} \) | −1.80 \[28\] |
| \( \text{Th}^{4+} + 4e^- \) | \( \text{Th} \) | −1.90 \[28\] |
| \( \text{K}^+ + e^- \) | \( \text{K} \) | −2.93 \[29\] |

In this work, a copper anode is used to provide \( \text{Cu}^{2+} \) ions to the electrolyte. For \( \text{Cu}^{2+} \) ions reducing at the cathode, the system will have \( E_{\text{cell}}^0 = 0 \) V. Thus, to drive the reaction and overcome energy loss mechanisms in the system, the electrodes are kept at a potential difference of 0.3 V \[30\].

It has been shown that applying a time-varying potential difference between the electrodes can have several benefits compared to a constant potential difference \[31\]. During electroplating, the region of the electrolyte at the surface of the cathode becomes depleted of \( \text{Cu}^{2+} \) relative to the bulk electrolyte. This slows down the rate of electroplating and affects the properties of the deposited copper \[31\]. The waveform of the time-varying potential difference allows this region to be replenished by allowing diffusion from the bulk electrolyte when no voltage is applied and by reintroducing more ions from the surface during the reverse-voltage part of the waveform. Also, differences in current density can arise due to differences in the distance between the anode and cathode surfaces (e.g. a surface rough point). High current density regions of the electrolyte are more depleted of \( \text{Cu}^{2+} \) than lower density regions. When no voltage is applied, ions can diffuse between two such regions and thus lead to a more uniform overall current density, while the reverse-voltage part of the waveform prevents a thick layer forming in the high current density regions \[31\]; both effects promote more uniform growth of the electroplated copper layer. The reversing of polarity also allows for release of contaminant ions that may have been entrapped during the high mass transport portion of the forward plating.

The waveform for the electroplating is shown in Fig. 5. Note that while the potential is applied it is potentiostatic at a level that favors the oxidation/reduction of copper.

![Figure 5: Waveform used in the electroplating. The negative terminal was attached to the cathode.](image)

### 4. Scale Model

The copper electroplating procedure described in the previous section is a well-established and successful method that has existed for over a decade \[23\]. However, this method must meet fairly
rigid operational conditions to produce optimal material. Failure to meet these conditions can not only produce copper of poor radiopurity but often results in deposits with poor physical properties as well. For NEWS-G the initial loading of copper into solution would need to be generated from an initial electropolishing step, because commercially available copper sulfate is not sufficiently pure. However, for traditional electroforming, the amount of copper required in the electrolyte is too great to achieve through electropolishing. As a result, the plating conditions for the NEWS-G hemispheres required a major deviation in the concentration of copper sulfate (CuSO\(_4\)) in the electrolyte.

Not all parameters have a well-studied effect on growth, especially when multiple parameters are outside of their established optimal operating ranges. Prior experience has shown that electrolyte with a low copper-ion concentration can produce dendritic copper deposition. In the absence of accurate deposition models it was necessary to run a scaled experiment prior to plating the full-sized \(\varnothing140\) cm sphere underground in Laboratoire Souterrain de Modane (LSM). Key growth parameters were identified and an experiment was designed based on those that could be adjusted \textit{in situ} at LSM and projected onto a scale model.

The key independent and adjustable variables were determined to be the concentration of copper and overall conductivity of the electrolyte, and the current based on the limiting set of voltage conditions. Control of the CuSO\(_4\) concentration is limited by the amount of copper that can be dissolved during an initial electropolishing step, which serves two purposes: a) expose the underlying bulk crystal structure to prepare the copper surface for electroplating; and b) load the electrolyte with copper. During this step, the \(\varnothing140\) cm hemisphere will act as the anode and careful control of the potential is not as important, whereas subsequently copper will be plated to the \(\varnothing140\) cm hemisphere which will then be serving as the cathode. During the latter step, the voltage control and deposition rate are critical. As a result, establishing how the plating responds to small changes in CuSO\(_4\) is crucial. As such, three variations of CuSO\(_4\) concentration, three conductivities, and three voltage settings were identified for experimentation on the scale model.

A stainless steel spherical float with a diameter of 30 cm was cut in half and used as a stand-in for the full-scale \(\varnothing140\) cm copper hemisphere. A smaller hemisphere was machined from aluminum and plated with copper to serve as the anode after the initial electropolishing step. Figure 6 shows the experimental setup of the scale model. Although the transport dynamics involved are not fully understood, previous experience has shown that the electrode gap (path length) has an effect on plating, regardless of CuSO\(_4\) concentration and conductivity. So, while the spacing between the two electrodes was scaled, the impedance needed to be matched to that of the full-scale setup. This required the electrolyte conductivity to be reduced to compensate for the reduced electrode spacing in the model.

A bath, shown in Fig. 6(a) and Fig. 6(b), was designed to hold and stabilize the stainless steel hemisphere, and several iterations of plating were performed to cycle through the plating variations and determine the optimal electroplating conditions. Based on these trials, the parameters chosen for plating copper onto the full-scale hemispheres are a CuSO\(_4\) concentration of 0.03 M, a conductivity of 91.9 mS/cm (corresponding to a full-scale conductivity of 300 mS/cm), and a potential of 0.35 V. Using these parameters, the estimated time to electroplate each full-scale hemisphere is \(\sim8\) days.
Figure 7: A detector hemisphere following (a) initial cleaning with detergent and (b) chemical etching with an acidified hydrogen peroxide solution.

to attain a thickness of 500 m. The resulting growth for the small-scale model is shown in Fig. 6(c) and Fig. 6(d).

5. Electroplating NEWS-G Detector

The electroplating was conducted at LSM at a depth of 4800 m water equivalent to reduce cosmogenic activation. The detector outer shell is comprised of two $\phi 140$ cm hemispheres, produced by a spinning technique using 4N copper. The result after cleaning with commercial detergent is shown in Figure 7(a). The hemispheres were then sanded to produce a smooth surface and subsequently chemically etched using an acidified hydrogen peroxide solution [32]. The result of this preparation is shown in Figure 7(b).

A smaller 4N copper hemisphere was produced to act as the anode for electroplating and was cleaned in the same way as the detector hemispheres. It was suspended inside the detector, separated by an electrolyte comprised of deionized water (18 Mohm), Optima® grade sulphuric acid (Fisher Scientific), and copper sulphate produced by a previous electroplating. A pump provided mechanical mixing with a filter removing particulates greater than 1 µm in size from the electrolyte. The anode and cathode were connected to a pulse-reverse power supply (Dynatronix, Amery, WI, USA), which could supply up to 80 A. The whole set-up was contained in a temporary purpose-constructed cleanroom to prevent particulates entering the electrolyte and subsequently providing nucleation sites for nodule-like copper growth [33]. The setup is shown in Fig. 8.

Prior to electroplating, each hemisphere was electropolished to remove a layer of material from the surface. This exposes the underlying crystalline structure and provides an ultraclean surface prior to deposition. Furthermore, this process enhances the amount of Cu$^{2+}$ in the electrolyte. A higher voltage was used for this process to extract all species from the surface. During electropolishing, (21.2 ± 0.1) µm and (28.2 ± 0.1) µm were removed from the first and second detector hemispheres, respectively. This was estimated from the integrated current and Eq. 3, assuming uniform polishing. Following this process, the electrolyte circulated through the filter for several days prior to electroplating to remove particulates released from the copper surface.

The electroplating procedure used the reverse-pulse plating waveform shown in Fig. 4. The current and voltage were monitored throughout, and the conductivity and temperature were recorded using a HACH inductive conductivity sensor. Electroplating continued for a total of 19.8 days and
21.0 days for the first and second hemispheres, respectively. The process took longer than estimated based on the small-scale experiments due to power supply current limitations, plating at slightly lower potential and a slightly lower electrolyte conductivity. The process was only interrupted for short periods to perform checks or due to power outages. The thickness of the deposited layer, which is shown as a function of time in Fig. 9 was estimated from the integrated current assuming a uniform deposition. Total copper thicknesses of $(502.1 \pm 0.2) \mu m$ and $(539.5 \pm 0.2) \mu m$ were plated onto the first and second detector hemispheres, respectively. The achieved plating rate corresponds to approximately $1.3 \text{ cm/year}$. A photo of the finished plating is shown in Fig. 10.

After removing the hemispheres from the setup, they were rinsed with deionized water and the surface passivated with a 1% citric-acid solution to prevent surface oxidation [32].

6. Radioisotope Assay Results

Samples of the electroplated copper were used to assess its $^{238}\text{U}$ and $^{232}\text{Th}$ concentrations. Samples were taken from copper plated on the stainless steel ring, shown in Fig. 8 to avoid damaging the detector cladding. These samples originate from near the electrolyte-air interface and from the stainless steel surface; thus, they represent a worst case scenario with respect to contamination. The samples collected from each hemisphere are shown in Fig. 11.

The samples were shipped to Pacific Northwest National Laboratory and analysed using ICP-MS following the methods described in Refs. [6, 16]. The results are summarized in Table 3 along with representative examples of electroformed and commercially sourced (machined) copper. A substantial improvement over the latter is observed, with radiopurity levels comparable to previously measured electroformed copper. The measurement sensitivity for the two hemispheres is limited by the mass of the available samples.

7. Summary

The NEWS-G collaboration has utilized recent advances in high-purity copper electroforming to produce a layer of copper on the inner surface of the $\Theta 140 \text{ cm}$ detector. This layer will act as a shield to mitigate background from $^{210}\text{Pb}$ in the bulk of the detector’s commercially sourced 4N copper. This is the largest surface to be plated with ultra-radiopure copper in an underground laboratory. This operation has demonstrated the feasibility of plating onto the surface of a large hemisphere. The radiopurity
of the plated copper was assessed using ICP-MS and found to be comparable to other electroformed copper. A copper deposition rate of approximately 1.3 cm/year was achieved, which is promising for fabrication of a fully electroformed copper sphere in the future.

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Table 3: ICP-MS results for $^{238}$U and $^{232}$Th contamination in samples of the electroplated copper layer, along with representative examples of electroformed and commercially sourced copper [17]. These are quoted as 68% upper confidence limits, where the measurement sensitivity was limited by the available sample mass.

| Sample   | Weight [g] | $^{232}$Th [µBq/kg] | $^{238}$U [µBq/kg] |
|----------|------------|---------------------|---------------------|
| Cl0100 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              |

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