1. Introduction

The NEWS-G collaboration [1] uses the spherical proportional counter [2], a novel spherical gaseous detector, in a direct search for low-mass Dark Matter (DM) particles. The detector comprises a grounded outer metallic spherical shell as the cathode and a centrally-located spherical anode at high voltage, while the volume in-between is filled with gas. Figure 1 shows a schematic of the detector. In the ideal case, electric field \( E(r) \) in the volume as a function of the radius \( r \) is given by,

\[
E(r) = \frac{V_0}{r^2} \frac{r_ar_c}{r_c - r_a} \hat{r},
\]

(1)

where \( V_0 \) is the anode voltage and \( r_a, r_c \) are the anode and cathode radii, respectively. Electrons produced by ionisation in the gas volume drift until arriving within micrometers of the anode where the electric field reaches \( \mathcal{O}(10 \text{ kV cm}^{-1}) \) and the electrons have sufficient energy to produce further ionisation. The electrons produced in the avalanche arrive at the anode almost immediately, however, the ions take \( \mathcal{O}(1 \text{ s}) \) to reach the cathode due to their reduced mobility. The majority of the measured signal is induced by the ions drifting towards the cathode. A detailed description of the detector is given in Ref. [4].

The spherical proportional counter offers many advantages which make its use attractive in a rare event experiments searching for dark matter. Compared to parallel plate and cylindrical geometry detectors, a sphere has the lowest capacitance allowing low electronic noise. This facilitates low energy thresholds which have been demonstrated previously, for example in Ref. [5]. For a dark matter experiment in an underground laboratory housed in appropriate shielding one of the main sources of background signals is nuclear disintegration in the construction materials of the detector. A sphere also has the smallest surface area to volume ratio, so this background is limited by the intrinsic low surface area of the spherical proportional
counter. Careful selection of radio-pure materials further reduces this background. The variation of the electric field with radius allows background interactions occurring near the surface to be discriminated against through pulse-shape analysis.

2. Background Rejection and Gas Quality

As shown by Equation 1, the electric field in the gas volume varies as the inverse of the radius squared. Due to the weaker electric field, primary electrons generated by ionisation at larger radii will undergo more diffusion. The increased diffusion in the primary electrons arriving at the anode relates to an increase in the rise time of the measured current pulse, allowing fiducialisation of the detector volume based on radius of interaction. Figure 2 shows the rise time of recorded pulses versus their amplitude for a spherical proportional counter, using an $^{55}$Fe source. $^{55}$Fe decays via electron capture to $^{55}$Mn and a 5.9 keV X-ray is released. X-rays from the source interacting through the gas volume are accumulated in region 1, with the rise time of the pulse dependent on the interaction radius of the X-ray. Signals originating from interactions near the cathode, region 2, may be rejected based on their larger rise time, which is where the majority of background processes occur. Particles which leave ionisation tracks may also be discriminated against using the rise time. Due to the spatial extent of primary electrons there will be a wider window of arrival times for the individual electrons to the anode, resulting in an increase in the rise time, demonstrated with the cosmic muons in region 3.

However, the low electric field for large radii can have a negative impact on detector operation in the presence gas contaminants. There is a greater probability of electron attachment by electronegative molecules such as oxygen and water. In the presence of such contaminants there is a reduction of primary electrons reaching the anode for amplification from ionisations generated at larger radii, which is observed as a reduction in amplitude for higher rise time signals. This reduces the ability of the detector to distinguish background events and also degrades the energy resolution. As a result the detector vacuum tightness and gas purity are of paramount importance.
Figure 2. Pulse rise time versus amplitude measured with a spherical proportional counter filled with He:Ar:CH$_4$ (51.7%:46.0%:2.3%). An Oxisorb filter was used to remove impurities from the gas. Region (1) are interactions of 5.9 keV X-rays from an $^{55}$Fe source occurring in the gas volume, (2) interactions occurring near the surface of the cathode and (3) are the interactions of cosmic muons in the gas, which are at higher rise times as they leave an extended track of ionisation.

3. Gas Purification

An initial study of the effect of electron attachment was carried out using a 30 cm diameter spherical proportional counter. Gas bottles with a minimum purity of 99.995% were used. To ensure that oxygen was not entering the detector through vacuum leaks the vacuum tightness of the gas system was improved to the level of approximately $10^{-5}$ mbar l s$^{-1}$. Due to the persistence of electron attachment effects the gas was filtered during its insertion into the detector. For this purpose a Messer Oxisorb $^1$ was used to remove oxygen and water impurities. Two measurements were carried out with a detector filled with 600 mbar of He:CH$_4$ (90%:10%), one without using the Oxisorb and one with. The attenuation length of the X-ray from the $^{55}$Fe source installed in the detector was approximately 30 m in this gas $^6$ and so is expected to interact throughout the entire gas volume.

Figure 3 shows the recorded pulse amplitude for the 5.9 keV photons measured with a spherical proportional counter filled with filtered and unfiltered gas. A selection on rise time has been applied to keep the volume interactions only (region 1 in Figure 2). Compared to the case where no filter was used, when the Oxisorb was used the recorded resolution, $\sigma$, was improved from 21.3 ± 0.7% to 9.4 ± 0.3%.

While this filter proved successful at removing impurities to a sufficient level to minimise electron attachment in the 30 cm diameter detector, it is technically challenging to use for larger detectors due to the low flow rate required for efficient filtering. Further tests were conducted with a SAES MicroTorr Purifier (MC700 902-F $^2$) commercial getter. This allows a maximum flow rate of approximately seven times higher than the Oxisorb and also intrinsically more efficient filtering $^3$. This was incorporated into the gas system for a 60 cm diameter spherical proportional

1 Messer Group GmbH, Gahlingspfad 31, 47803 Krefeld, Germany
2 SAES Pure Gas, 4175 Santa Fe Rd., San Luis Obispo, CA 93401, USA
3 MC700 902-F specifications may be found at http://www.saespuregas.com/Library/purifier_specifications/902_Media_Specification.pdf
Figure 3. Comparison of pulse amplitude measured for the 5.9 keV X-rays from an $^{55}$Fe source using a spherical proportional counter in the case when the gas was passed through an Oxisorb$^1$ filter and when it was not. Filtering improved the resolution by a factor of 2.3. The second peak at lower energy is the 1.49 keV X-ray from the fluorescence of the $^{55}$Fe source’s aluminium casing.

counter. Although the absence of reduced amplitudes at higher rise times indicated that this efficiently filtered oxygen and water vapor from the gas, it was found that large amounts of $^{222}$Rn gas were introduced. This has previously been reported by several experiments [7, 8]. This has also been found for the Oxisorb [9]. While the $\alpha$-particle released by the $^{222}$Rn decay can be rejected through pulse shape analysis, $\beta$-particles from the decay of daughter isotopes can contribute to the experimental background of the detector reducing the sensitivity to DM. Work is ongoing to incorporate a carbon filter, inserted after the oxygen filter in the gas system to remove any emanated $^{222}$Rn.

It is important to note that in addition to gas impurities, the electric field strength is key to the effect of electron attachment in the detector. Increasing the magnitude of the electric field at large radii in the spherical proportional counter is also an active area of investigation through the use of a multi-anode module, ACHINOS [10].

4. Copper Purity

With one spherical proportional counter, SEDINE, currently producing results in the search for low-mass DM candidates [4], the NEWS-G collaboration is constructing the next generation detector, NEWS-G@SNOLAB, schematically shown in Figure 4. NEWS-G@SNOLAB, with a diameter of 135 cm, will have over ten times the volume of SEDINE in an effort to increase its sensitivity and will also be constructed using 4N (99.99% pure) Aurubis$^4$ copper to reduce the background introduced by nuclear disintegration of contaminants in the detector material. Copper has no long-lived radioisotopes making it an ideal construction material for a low-background experiment. However, it was recently demonstrated that even such pure copper contained unacceptable amounts of $^{210}$Po and $^{210}$Pb [11]. From the same decay chain as $^{210}$Rn, these isotopes negatively impact the sensitivity of the experiment. The effect of the additional background on the count rate in the NEWS-G@SNOLAB detector was simulated using the

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$^1$ Oxisorb

$^4$ Aurubis AG, Hovestrasse 50, 20539 Hamburg, Germany
Geant4 toolkit [12]. It was found that the decays of $^{210}$Pb and $^{210}$Bi contributed 4.6 dru (1 dru = 1 count/keV/kg/day) below 1 keV, which is approximately an order of magnitude larger than other background contributions in this energy range. For this reason it was decided to use electroplating to add a cladding-type layer of ultra-pure copper to the cathode surface. Other experiments have also used electroplating and electroforming to minimise radioisotope contamination in copper [13], using a established techniques, e.g. Refs. [14, 15]. In electroplating, an applied voltage between and cathode and an anode induces an electrolytic current through an electrolyte solution. This facilitates reduction reactions at the cathode which result in the deposition of ions from the electrolyte onto the cathode’s surface. The mass deposited $M$ over a given time $t$ is a function of the current $I$, and is given by Faraday’s laws of electrolysis,

$$M = \frac{m_r}{zF} \int I(t) dt,$$

where $m_r$ is the molecular mass of the electroplated molecule, $z$ the number of electrons required for the reduction and $F$ is the Faraday constant, defined as the charge of a mole of electrons $eN_A$ with $e$ the electron charge and $N_A$ the Avogadro number.

In any electrolyte there will be several species of ion available to electroplate to the cathode. The high reduction potential of copper (+0.34 V) means that it will preferentially electroplate over impurities with lower reduction potentials such as lead (−0.13 V), uranium (−1.80 V) and thorium (−1.90 V). By carefully choosing the applied voltage between the anode and cathode only copper (and molecules with a higher reduction potential) will be electroplated. In this way, an ultra-pure layer of copper may be formed on a surface.

A 500 µm layer of ultra-pure copper plated onto the inner cathode surface of the NEWS-G@SNOLAB detector, is estimated from a Geant4 simulations to reduce the background to 2.0 dru below 1 keV.

5. Electroplating of NEWS-G@SNOLAB

The electroplating was carried out in the underground laboratory in Modane, Laboratoire Souterrain de Modane (LSM) [16]. The detector outer shell is comprised of two hemispheres. These were first cleaned with commercial detergent and then sanded to remove raised portions of copper. The surface was then chemically etched using an acidified hydron peroxide solution [15].

To act as the anode for electroplating a smaller hemisphere of copper was prepared. This was suspended inside the detector hemisphere, separated by an electrolyte comprising deionised water and sulphuric acid. During the electroplating process the anode would be oxidised to increase the...
concentration of copper ions in the electrolyte. A pump installed to provide mechanical mixing while an attached filter removes particulates greater than 1 \( \mu \text{m} \) in size from the electrolyte. The set-up is shown in Figure 5.

The surface was first electropolished to ensure the surface was smooth before beginning electroplating and to expose the underlying crystal structure. This process is the opposite of electroplating and removes copper from the detector hemisphere. \((21.2 \pm 0.1) \mu \text{m}\) and \((28.2 \pm 0.1) \mu \text{m}\) were removed from each of the detector hemispheres. This process also increases the copper sulphate concentration of the electrolyte.

To perform the electroplating a reverse pulse voltage application was used, with the pulse shape shown in Figure 6. This has been demonstrated to improve the properties of the deposited copper, for example see Ref. [17] for a review of this technique. The potential difference used between the anode and the cathode for electroplating was 0.3 V, which is an established value for electroplating pure copper. Electroplating continued for approximately 15 days for each hemisphere and the current was recorded throughout. With the assumption that the deposited layer was uniform in thickness, Equation 2 to be used to estimate that \((502.1 \pm 0.2) \mu \text{m}\) and \((539.5 \pm 0.2) \mu \text{m}\) of copper were plated onto the surface. It is important to note that the quoted uncertainty does not include any uncertainty due to the roughness of the surface. This equates to a plating rate of approximately 1.3 cm year\(^{-1}\).
The surface was then rinsed with water and the surface passivated with a 1% citric acid solution [15]. Currently the detector is being assembled using electron beam welding in preparation for first operation, which is planned for May 2019 in LSM. Analysis of samples of copper and electrolyte collected during the operation aim to assess the achieved radio-purity of the electroplated layer and are currently ongoing.

6. Future Radio-Pure Spherical Proportional Counters

To achieve even greater detection sensitivity for low mass DM candidates, future NEWS-G projects must have even greater radio-purity to further reduce backgrounds. Two main avenues exist for this: Commercial 6N (99.9999%) pure copper or an electroformed sphere, each having their own advantages. 6N copper is commercially available to purchase, greatly simplifying manufacturing a detector. However, the manufacturing process required to produce a sphere from this copper may introduce further contaminants. Electroforming a spherical detector directly has not been attempted previously adding to the technical complexity of the project. However, the electroplating of the NEWS-G@SNOLAB sphere demonstrates the possibility to perform electroplating on spherical surfaces and has addressed many difficulties which could be expected in electroforming a complete sphere.

7. Conclusion

As with any proportional counter the spherical proportional counter is sensitive to impurities in the gas that can cause electron attachment. The pulse rise time is an established back ground rejection parameter but its usefulness is impaired when electronegative contaminant gases are present due to their increased effect on primary electrons generated in the low electric field at high radii. Two oxygen filters have been tested and found to be sufficient at removing such impurities. However, these filters were also found, as previously reported, to emanate unacceptable amounts of $^{222}\text{Rn}$ gas. This would contribute a substantial experimental background to the DM search carried out by the NEWS-G collaboration. Efforts to further improve on this are currently on-going.

Another important factor in background reduction is the construction materials of the detector. For the newest spherical proportional counter to be employed by the collaboration, 4N (99.99%) pure copper has been selected. It has been further electroplated with a cladding-type layer of ultra-pure copper, which is expected to further reduce background contributions from the copper by a factor of between 2 and 3. Chemical assessment of the electroplated copper and the electrolyte solution used are ongoing to assess the reduction in contaminants. The electroplating of this detector has demonstrated the feasibility of this technique to be used in plating large hemisphere surfaces. This achieved a copper deposition rate of approximately $1.3 \text{ cm year}^{-1}$, giving promise for the future prospect of electroforming an intact copper sphere to be used as a spherical proportional counter in a direct DM search. Alternatively, a future low background spherical proportional counter using commercially available 6N (99.9999% pure) copper, which although not as radio-pure as electroforming an intact sphere, is still a significant advancement in radio-purity.

8. References

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