Confronting the Radiological Screening Challenges for Next-generation Rare-event Detectors

XinRan Liu[a)]

School of Physics & Astronomy, The University of Edinburgh, Edinburgh, United Kingdom

[a)] *Corresponding author: x.liu@ed.ac.uk*

Abstract. Over the past few decades, the scale and mass of rare-event search experiments have increased by several orders of magnitude. To maintain background-free large fiducial-volume searches, the radiopurity requirements of the materials from which these devices are constructed have improved by similar factors. High-purity germanium spectroscopy has long-been the workhorse of material screening and selection, providing information on trace radioactive gamma-ray-emitting impurities in the bulk of materials. The next generation of direct dark matter and neutrinoless double beta decay experiments demand the development of additional assay techniques to provide a more complete understanding of the full uranium (U) and thorium (Th) decay chains, including knowledge of alpha-emitting surface depositions. In this proceeding I highlight the challenging radiopurity requirements for the next generation of rare-event search experiments, as well as the extensive UK-based material-assay infrastructure in place to address these demands. Where requirements exceed current capability, additional R&D is needed. I will summarise where this R&D is already underway across the UK.

INTRODUCTION

The discovery of the Higgs Boson completed the Standard Model of Particle Physics, the most rigorously tested model in history. Yet the Model does not include gravity, neutrino mass and oscillation, or the dark sector that is known to make up 95% of the Universe. Over the coming decade particle physics will transition to the exploration of physics beyond the Standard Model, tackling topics such as dark matter and neutrinoless double beta ($0\nu\beta\beta$) decay. It is widely anticipated that these will be the keys to unlocking a more fundamental understanding of Nature.

The field of direct detection of weakly interacting massive particles (WIMPs) has been dominated by dual-phase xenon time projection chambers (TPCs), but the radiopurity problem is faced by all experiments regardless of technology. The scale and scope of the dual-phase xenon TPC experiments have expanded rapidly over the past decades from the 3-kg liquid-xenon target of ZEPLIN-I [1] in the mid 2000s to 10 tonnes of liquid xenon deployed by the LUX-ZEPLIN experiment setting the most recent world-leading dark matter limit [2]. With each new generation of dark matter detector, there are significant increases in target mass and improvements to instrumentation and construction materials to meet the increasingly stringent radiopurity requirements.

Similar radiopurity challenges and demands are faced by experiments searching for neutrinoless double beta decay. Since neutrino experiments, such as Super-Kamiokande [3] and SNO [4], have observed and confirmed the phenomenon of neutrino oscillations, first postulated by Bruno Pontecorvo in 1957 [5, 6], neutrinos must also have non-zero mass. Neutrinos may therefore be either Dirac or Majorana fermions. In the Dirac case, the neutrino would have a distinct anti-neutrino counterpart, whereas, in the Majorana case the neutrino would be its own antiparticle.

The $0\nu\beta\beta$ decay is the most sensitive and perhaps the only practical way to probe whether neutrinos are Dirac or Majorana fermions and will provide complementary information to cosmology and oscillation experiments on determining the absolute neutrino mass and the mass hierarchy. The observation of $0\nu\beta\beta$ decay would provide unambiguous evidence of lepton number violation required by theories of leptogenesis and baryogenesis to explain the matter-antimatter asymmetry of the universe and is also a requirement of almost every single GUT model. The observation of $0\nu\beta\beta$ decay would therefore have a profound impact on the field of particle physics and beyond.

Given the broad similarity in radiopurity requirements for all rare-event search experiments, this paper will generalise and where necessary highlight experiment-specific demands. A large variety of experiments using different techniques have probed $0\nu\beta\beta$ half-life and searched for dark matter over the past decades, without observation, setting ever more stringent limits.

FUTURE BACKGROUND REQUIREMENTS

Successive experiments have relied on increasing detector mass to further increase detection sensitivity. For a given set of detector components, the detector background usually increases linearly with mass and exposure time; hence

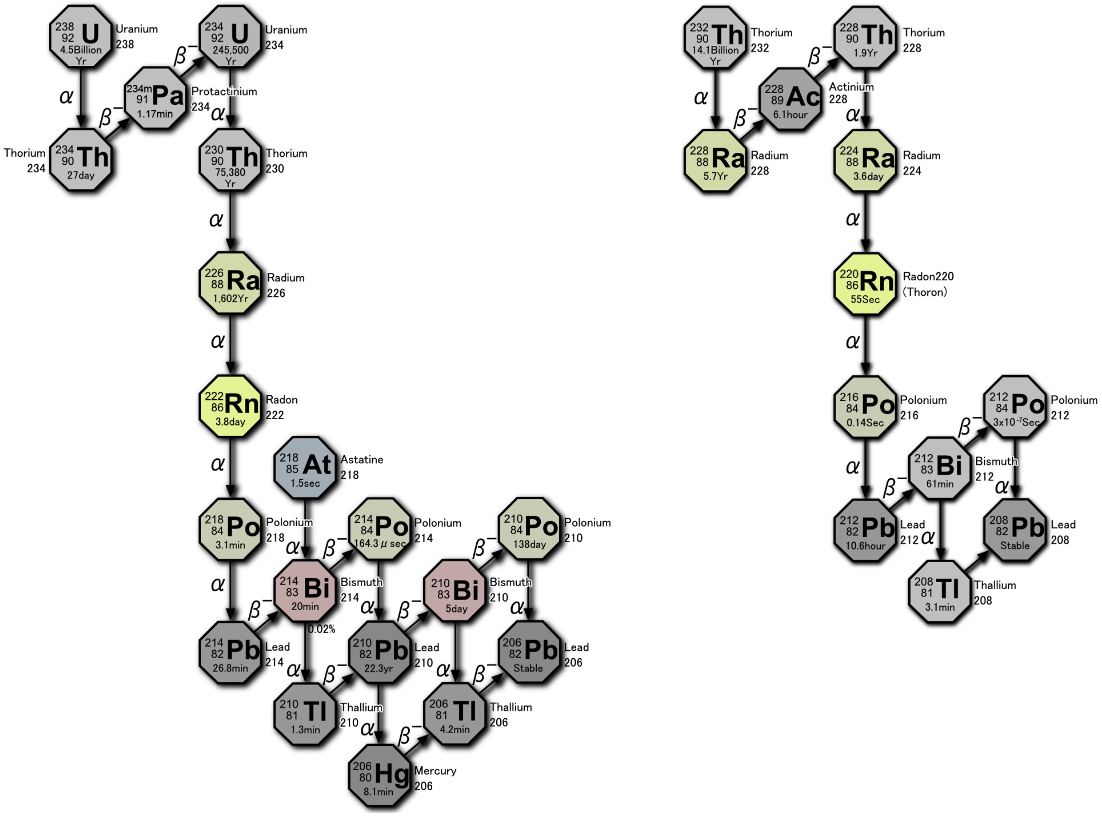

FIGURE 1. Left: The decay chain for ^{238}U [8]. **Right:** The decay chain for ^{232}Th [9].

the sensitivity of experiments is proportional to $\sqrt{Mt}$. It is, therefore, crucial to minimise background activity. The uranium and thorium decay chains, presented in Fig. 1, are the dominant backgrounds for any rare-event search experiments. In the case of $0\nu\beta\beta$-decay experiments, U and Th daughter isotopes ^{214}Bi and ^{208}Tl respectively are of particular concern. They have decay Q values ($Q_\beta(^{214}\mathrm{Bi})$ = 3.27 MeV and $Q_\beta(^{208}\mathrm{Tl})$ = 4.992 MeV) high enough to mimic a $0\nu\beta\beta$ event. The expected half-life sensitivity of any $0\nu\beta\beta$ can be approximated as [7]

$$T^{0\nu}_{1/2} \propto a \cdot \varepsilon \cdot \sqrt{\frac{Mt}{\Delta E \cdot N_\mathrm{B}}}, \tag{1}$$

where $T^{0\nu}_{1/2}$ is the half-life sensitivity to $0\nu\beta\beta$ in years, ε is the event detection and identification efficiency, a is the isotopic abundance of the $0\nu\beta\beta$ source isotope in the source mass, ΔE is the energy resolution, Mt is the total exposure of the experiment in kg yr, and N_B is the number of expected background events for the exposure.

The next generation of $0\nu\beta\beta$ experiments such as LEGEND, nEXO, and CUPID will have approximately an order of magnitude more detector mass to produce a sensitivity that exceeds the half-life of order 10^{28} years to completely explore the inverted mass hierarchy. Given the square-root dependence between the target sensitivities and the expected background events for the exposure, it is critical to keep radiopurity under control.

Initial considerations suggest the next generation (G3) of dark matter detector should comprise 50–90 tonnes of xenon. A G3 dark matter experiment will improve spin-independent WIMP-nucleon scattering cross-section sensitivity by more than an order of magnitude upon the LUX-ZEPLIN (LZ) and XENONnT experiments. This leap in sensitivity will explore the theoretically favoured parameter space down to the so-called "neutrino fog" that represents the limit for existing technology. If LZ has already made the first discovery, G3 is even more important, as it will be the instrument with which to first measure the properties of dark matter. For both dark matter and $0\nu\beta\beta$, the order of magnitude increase in detector mass results in the requirement for a similar order of magnitude improvement to existing assay capabilities.

FIGURE 2. Boulby UnderGround Screening (BUGS) Facility.

CURRENT UK ASSAY CAPABILITIES

Achieving the scientific goals of the next generation of rare-event search experiments is not simply a matter of scaling up existing technology. Widely recognised as one of the greatest unsolved challenges is how to meet the stringent radiopurity requirements needed to reach the proposed sensitivity. Current material-assay capabilities need to improve both in sensitivity and in throughput. Coordinated international efforts to cross-calibrate existing facilities and share best practise when establishing new laboratories is essential. Alongside improvements to individual detection methods, complementary techniques should be used in conjunction to build a complete understanding of the U and Th decay chains and the radioactivity from early to middle and late regions of the decay chain. The current and near-future material-assay capabilities for the UK are summarised in this section.

Boulby UnderGround Screening (BUGS) Facility

The BUGS facility is located within the Boulby Underground Laboratory situated 1070 m (2805 m w.e.) underground [10]. The laboratory has the lowest ambient radon levels of any deep underground facility at $\sim$2 Bq/m^3. BUGS is designed and operated as a class-1000 cleanroom with air conditioning to maintain a stable temperature, a nitrogen gas line to deliver purging gas for detectors, and a sample-preparation area. The cleanroom further minimises background variation as it reduces the level of contamination in the air. The facility was designed to become a centre of excellence for material assay and cleanliness, with the aim to bring together all current assay techniques into one location; see Fig. 2.

Radon-Removal System

Nitrogen is produced underground for cooling and flushing of detectors. Despite the low ambient levels of radon at Boulby, without mitigation radon would remain the dominant background for assay techniques being deployed at BUGS due to continued improvements to detector sensitivity. In fact, even underground generated nitrogen used as a purge would provide too large a radon background if untreated. A radon-removal system was therefore designed, based on the system developed by the SuperNEMO collaboration [11], and installed; see Fig. 3. Various carrier gases and activated charcoals were tested when first building the system to determine the optimum conditions for radon removal. The system is capable of removing radon from a carrier gas by up to 10 orders of magnitude depending on the gas.

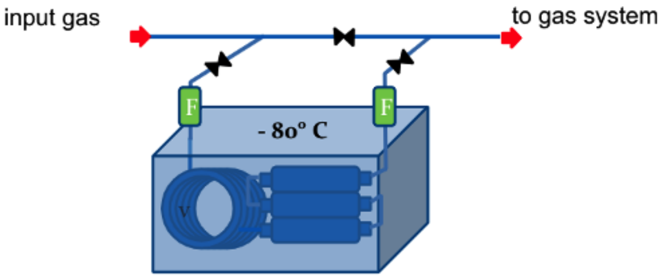

FIGURE 3. Schematic of the Boulby radon-removal system.

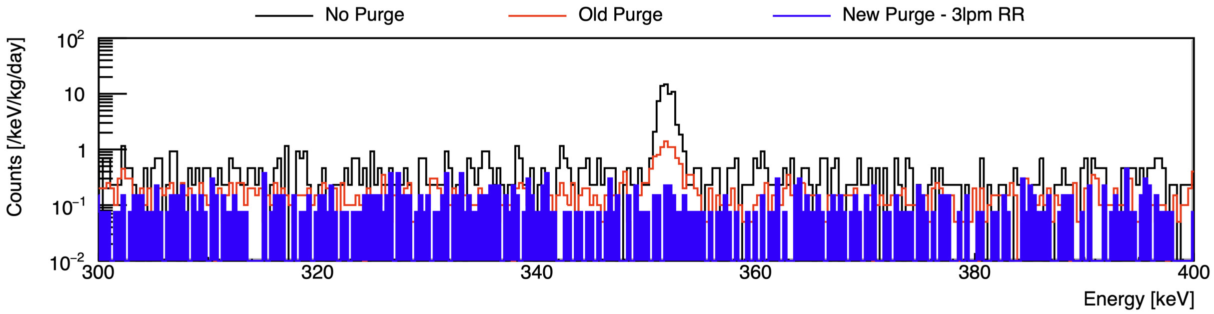

FIGURE 4. Background spectra of the Belmont detector with no purge (black) compared to with nitrogen purge (red) and with radon-reduced nitrogen purge (blue). The radon-reduction system has demonstrated background reductions by more than two orders of magnitude in the region of interest, in this case around the ^{214}Pb $\rightarrow$ ^{214}Bi energy at 352 keV.

The system, which uses activated charcoal cooled to -80°C, was installed to purify the nitrogen purge gas used to flush the BUGS germanium detectors, the results of which can be seen in Fig. 4. Due to the success of the system, two more radon-reduction systems are currently being commissioned and installed, each capable of delivering 30 slpm of continuous radon-free (< 20 µBq/m^3) nitrogen gas.

Gamma-ray Spectroscopy

The backbone of the BUGS facility is gamma-ray spectroscopy using high-purity germanium (HPGe) detectors. An ambitious programme to reboot and expand the UK underground-screening facility was launched in 2013 with the refurbishment of the existing p-type coaxial detector which was then renamed Lunehead. In 2016 an additional 3 specialty ultra-low-background detectors in purpose-built castles were added to the current class-1000 cleanroom. These were further supplemented with a specialty ultra-low-background SAGe well detector commissioned in 2021. The background summary shown in Table I demonstrates world-class detector backgrounds.

TABLE I. BUGS germanium detector background rates.

Detector	Relative Efficiency/ Type	Count Rate (kg^{-1} day^{-1}) 351 keV (^{214}Pb)	609 keV (^{214}Bi)	238 keV (^{212}Pb)	1461 keV (^{40}K)	2615 keV (^{208}Tl)
Roseberry	BE6530	0.15 (7)	0.15 (7)	0.8 (3)	0.8 (2)	0.2 (1)
Chaloner	BE5030	5 (1)	4 (1)	7 (1)	8.4 (14)	2.1 (5)
Belmont	160%	0.7 (2)	0.4 (1)	0.13 (6)	1.0 (2)	0.3 (1)
Merrybent	100%	2.5 (3)	1.8 (3)	0.3 (1)	1.9 (3)	0.8 (2)
Lunehead	100%	5.6 (5)	4.7 (4)	8.3 (5)	9.1 (6)	2.0 (3)
Lumpsey	SAGe-Well	1.1 (7)	1.3 (3)	1.1 (7)	1.7 (7)	0.7 (2)

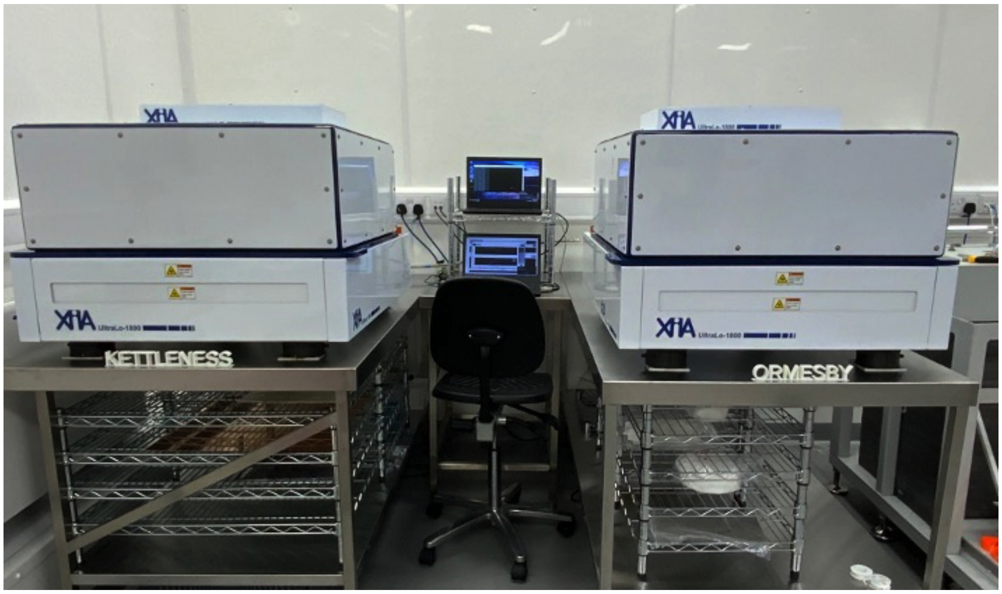

FIGURE 5. BUGS XIA UltraLo-1800 detectors Kettleness (left) and Ormesby (right).

TABLE II. BUGS XIA UltraLo-1800 detector backgrounds with sample tray.

Sample	Duration (hrs)	Alphas	Surface Area (cm^2)	Emissivity (α/khr/cm^2)	Activity (mBq/m^2)
Background (SS Tray)	168	342	1800	1.24 ± 0.07	6.88 ± 0.38
Background (PTFE Linear)	168	103	1800	0.38 ± 0.04	2.12 ± 0.22
Background (PNNL Copper)	168	13	707	0.13 ± 0.04	0.72 ± 0.22

Despite improvements in detector backgrounds, current BUGS detectors are only able to reach ∼100 µBq/kg levels of sensitivity. However, BUGS is currently acquiring a system to run multiple HPGe in coincidence, which would significantly improve existing capabilities.

Alpha Spectroscopy

The decays of radon and thoron lead to charged daughter atoms that can ‘plate out’ by electrostatic attraction onto nearby surfaces. Subsequent decays of these surface deposits are problematic in rare-event searches, an issue that is compounded by the possibility of the recoiling heavy ions implanting to depths of hundreds of nm, making removal challenging. Alpha particles emitted from component surfaces may illuminate neighbouring components containing nuclides with large (α, n) cross sections such as ^{19}F in PTFE.

Control of surface and bulk alpha-particle activity during material construction and assembly is vital and requires fast, accurate measurement to determine initial material contamination, exposure, and plate-out. The XIA UltraLo-1800 detector is the state-of-the-art surface-alpha detector with world-leading sensitivity to surface-alpha emission. Two XIA detectors have been installed in the BUGS facility, commissioned and fully operational since 2018, capable of measuring ^{210}Po surface contamination to 1 mBq/m^2 and bulk contamination to 30 mBq/kg.

Internal XIA studies show a 3× reduction in background rates as a result of being deep underground and free from cosmic events. Further reductions were made to the detector background with sample tray only by the addition of a 0.05 mm thick sheet of conductive-graphite filled PTFE (DW 105 by DeWAL) liner. The results are shown in Table II, where electroformed copper from Pacific Northwest National Laboratory (PNNL) has been shown to be an ideal material to be used as the sample tray. Most or all of the activity measured with the electroformed-copper liner is expected to be due to alpha emission from detector materials together with events from radon in the argon buffer gas required for operation. A radon-removal system will be installed to purify the input argon gas.

Radon-Measurement Facility

BUGS is currently constructing a radon-measurement facility with two ultra-low-background 80-L electrostatic radon detectors which have a background rate of ∼1 count per day [12]. The two detectors can be used independently or in parallel with a concentration line to enable the measurement of radon emanation from large gas volumes; see Fig. 6.

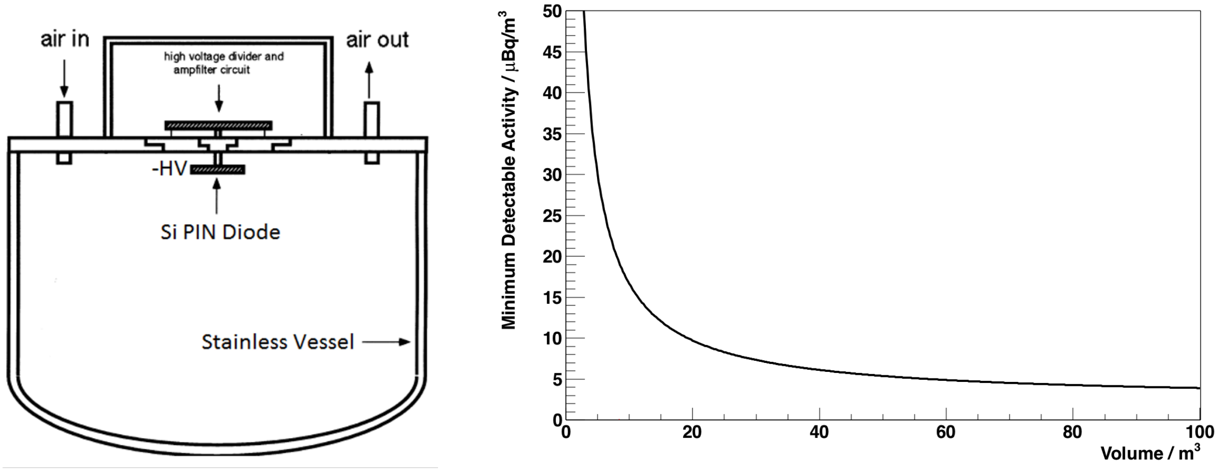

FIGURE 6. Left: Schematic of an 80-L electrostatic radon detector. **Right:** Preliminary estimate of radon concentration line minimum detectable activity as a function of gas volume measured

The new system is designed to offer increased measurement flexibility and sample measurement rates. A series of emanation chambers will be installed with the new facility ranging from 5, 10, 20 to 100 L with plans for larger chambers when needed. The new system is expected to achieve a conservative sensitivity of 5 μBq/m^3 for large gas volumes and ∼100 μBq per sample using the small emanation chambers.

Electroforming Copper

Copper is frequently used for rare-event search experiments. However, commercial copper can be contaminated by U and Th in the raw material as well as by cosmic muon spallation. Boulby Underground Laboratory plans to develop the capability to electroform copper underground. The work will be led by the NEWS-G collaboration with their ECuMe experience having developed this technique at SNOLAB. This capability will be highly complementary to the BUGS facility, the existing detectors, and future projects at Boulby Underground Laboratory.

Cleaning and Storage Facility

In conjunction with the rapid expansion of BUGS facility, plans are underway to establish a surface material-cleaning laboratory. The surface laboratory would provide a controlled environment and allow detailed study of how to best clean and keep clean various materials used in future rare event search experiments.

Inductively coupled plasma mass spectrometry (ICP-MS)

The UK particle physics ICP-MS capability is currently a state-of-the-art Agilent 8900 tandem-mass triple-quadrupole ICP-MS, optimised for extremely low detection limits, located at University College London (UCL) inside a dedicated class-1000 cleanroom. The system is fitted with an inert sample-introduction kit that includes a PFA nebulizer, 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 acid concentration in the sample introduced to the ICP-MS, including hydrofluoric acid [13].

The facility has the relevant infrastructure to provide reproducible high-throughput sensitivity at the parts-per-trillion (ppt) level for direct measurements of ^{238}U and ^{232}Th. The advantage of ICP-MS is it offers very fast sample-turn-around rate at 1–2 samples per day instead of per week or month in some cases. The existing facility will eventually move to Boulby's surface cleaning and storage facility to complete its screening capabilities.

Radon Concentration Line (RnCL)

The RnCL was a system developed by the SuperNEMO collaboration at UCL to measure the radon emanation rates of the demonstrator module located at the Modane Underground Laboratory [14]. The RnCL system essentially consists of a stainless steel trap containing ultra-radiopure activated carbon which is cooled to -50°C. The cooling significantly improves the charcoal's ability to capture radon. Large volumes of gas can be flowed through the trap where the radon is adsorbed. After the gas volume to be measured has passed through the charcoal trap, it is sealed and the concentrated radon sample can then be heated, releasing the radon, which can then be flushed into the electrostatic detector for measurement.

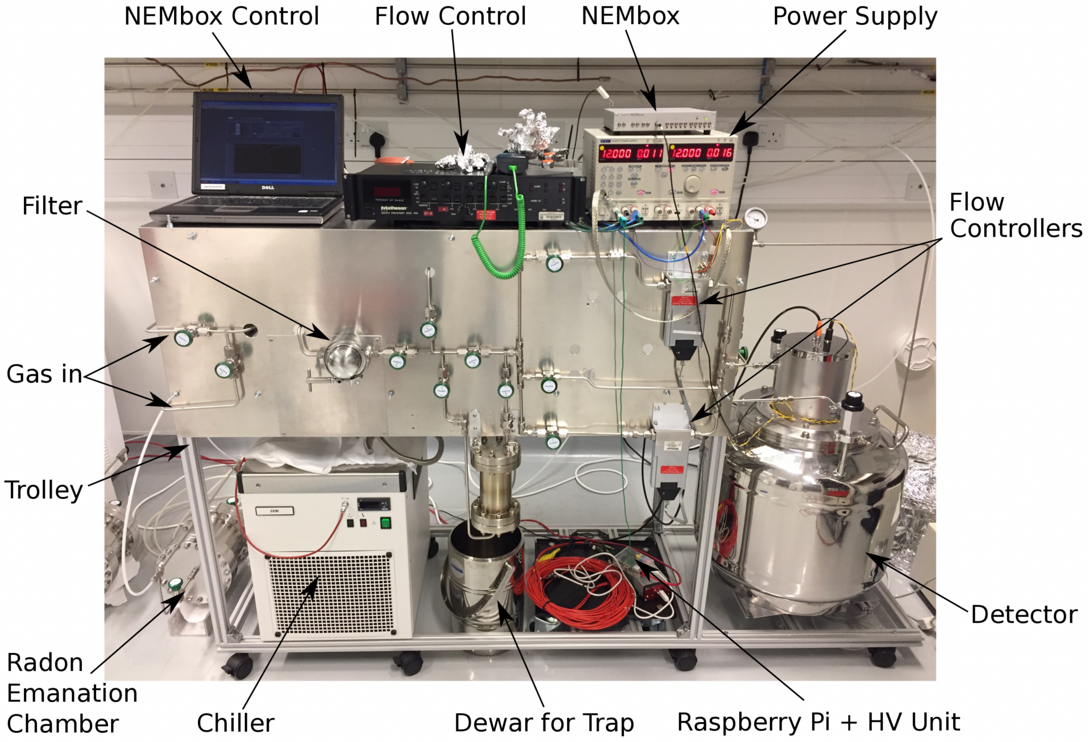

FIGURE 7. Radon concentration line.

Combining the RnCL with an electrostatic detector the system can reach sensitivities as low as 5 µBq/m^3 for large gas volumes as shown in Fig. 6. There is however diminishing returns as the volume of gas required to improve the minimum detectable activity (MDA) increases exponentially.

Cold Radon Emanation Facility (CREF)

The Cold Radon Emanation Facility (CREF) is currently being commissioned at Rutherford Appleton Laboratory in the UK as shown in Fig. 8. The goal of the system is to deliver a world-leading sensitivity below 0.1 µBq/kg for ^{222}Rn emanated from materials at temperatures of relevance to next-generation rare-event searches. The facility inherits heavily from existing radon-emanation assay systems by utilising a radon-concentration line and a silico-pin-diode alpha detector as detailed in the section on the radon measurement facility.

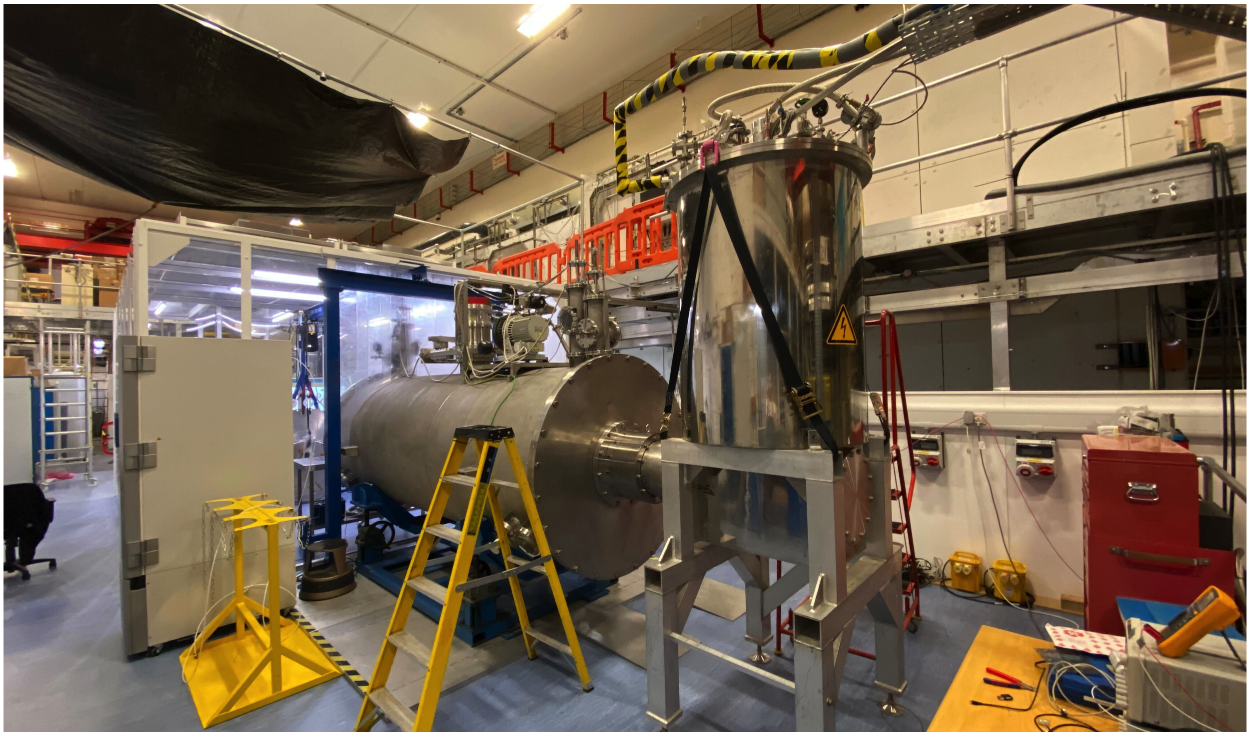

FIGURE 8. Cold Radon Emanation Facility at the Rutherford Appleton Laboratory.

Additionally, CREF is designed to be operated with the emanation chambers cooled to LAr or LN_2 temperatures for low-temperature measurements. Finally, CREF incorporates a large, 200-L chamber, operating within a 500-L cryogenic vessel, that can be cooled and stabilised at temperatures down to $\sim$77 K. This allows measurements of 'as built' detector components and to establish their rate of emanation as a function of temperature.

CONCLUSION

The success of the next generation of rare-event search experiments depends explicitly on achieving stringent radiopurity requirements. To meet this demand, an order of magnitude improvement on all our existing assay techniques is necessary along with a coordinated international effort to cross-calibrate existing facilities.

The UK has already started an ambitious programme to upgrade existing assay facilities with BUGS at the core as the centre for material screening and cleanliness. The radon-removal system can purify nitrogen gas to 20 μBq/m^3 which has resulted in factor-150 reductions to HPGE backgrounds as a result of radon decay daughters. The Belmont detector has demonstrated the capability to reach $\sim$100 μBq/kg levels, and new multi-HPGe-in-coincidence systems will further improve on existing sensitivity. The radon facility can be expected to reach 5 μBq/m^3 for large gas volumes and 100 μBq/m^3 per sample for emanation. The XIA detectors have demonstrated world-class sensitivity reaching of order 1 mBq/m^2 for surface ^{210}Po and 30 mBq/kg for bulk contamination.

UK low-background science is working closely with industrial partners to continually improve commercial detector sensitivity in order increase sample measurement throughput. BUGS is complemented by the Cold Radon Emanation Facility, which will provide an unprecedented understanding of radon emanation at temperatures and conditions relevant to rare-event searches.

ACKNOWLEDGMENTS

The author would like to thank Emma Meehan and Paul Scovell from the Boulby Underground Laboratory, Jim Dobson and Emily Perry from UCL, and Patrick Knight from University of Birmingham for providing key information and images for this publication.

REFERENCES

1. G. J. Alner *et al.* (UK Dark Matter Collaboration), "First limits on nuclear recoil events from the ZEPLIN I galactic dark matter detector," Astroparticle Physics **23**, 444–462 (2005).
2. J. Aalbers *et al.* (LZ Collaboration), "First dark matter search results from the LUX-ZEPLIN (LZ) experiment," (2022), arXiv:2207.03764 [hep-ex].
3. Y. Fukuda *et al.* (Super-Kamiokande Collaboration), "Evidence for oscillation of atmospheric neutrinos," Phys. Rev. Lett. **81**, 1562–1567 (1998), arXiv:hep-ex:9807003.
4. Q. R. Ahmad *et al.* (SNO Collaboration), "Measurement of the rate of $\nu_e + d \to p + p + e^-$ interactions produced by ^{8}B solar neutrinos at the Sudbury Neutrino Observatory," Phys. Rev. Lett. **87**, 071301 (2001).
5. B. Pontecorvo, "Mesonium and antimesonium," Soviet Journal of Experimental and Theoretical Physics **6**, 429 (1958).
6. B. Pontecorvo, "Inverse beta processes and nonconservation of lepton charge," Soviet Journal of Experimental and Theoretical Physics **7**, 172–173 (1958).
7. F. T. Avignone, S. R. Elliott, and J. Engel, "Double beta decay, Majorana neutrinos, and neutrino mass," Rev. Mod. Phys. **80**, 481–516 (2008).
8. Tosaka, "File:decay chain(4n+2, uranium series).png — wikimedia commons, the free media repository," `https://commons.wikimedia.org/w/index.php?title=File:Decay_chain(4n\%2B2,_Uranium_series).PNG&oldid=526952474` (2021), licenced under `https://creativecommons.org/licenses/by/3.0/legalcode`; online; accessed 15-August-2022].
9. Tosaka, "File:decay chain(4n,thorium series).png — wikimedia commons, the free media repository," `https://commons.wikimedia.org/w/index.php?title=File:Decay_chain(4n,Thorium_series).PNG&oldid=510909841` (2020), licenced under `https://creativecommons.org/licenses/by/3.0/legalcode`; online; accessed 15-August-2022].
10. H. Araújo, J. Blockley, C. Bungau, M. Carson, H. Chagani, E. Daw, B. Edwards, C. Ghag, E. Korolkova, V. Kudryavtsev, P. Lightfoot, A. Lindote, I. Liubarsky, R. Lüscher, P. Majewski, K. Mavrokoridis, J. McMillan, A. Murphy, S. Paling, J. Pinto da Cunha, R. Preece, M. Robinson, N. Smith, P. Smith, N. Spooner, T. Sumner, R. Walker, H. Wang, and J. White, "Measurements of neutrons produced by high-energy muons at the Boulby Underground Laboratory," Astroparticle Physics **29**, 471–481 (2008), arXiv:0805.3110 [hep-ex].
11. X. R. Liu, "UK low-background infrastructure for delivering SuperNEMO," (2015), arXiv:1504.08335 [physics.ins-det].
12. K. Hosokawa, A. Murata, Y. Nakano, Y. Onishi, H. Sekiya, Y. Takeuchi, and S. Tasaka, "Development of a high-sensitivity 80 L radon detector for purified gases," Progress of Theoretical and Experimental Physics **2015**, 033H01 (2015).
13. J. Dobson, "Low-background radioassay facilities," `https://www.hep.ucl.ac.uk/darkMatter/radioassay.shtml` (2020), [Online; accessed 15-August-2022].
14. X. R. Liu (for the SuperNEMO Collaboration), "Radon mitigation strategy and results for the SuperNEMO experiment," in *XXVII International Conference on Neutrino Physics and Astrophysics (Neutrino2016) 4–9 July 2016, United Kingdom*, Journal of Physics: Conference Series, Vol. 888 (IOP Publishing, 2017) p. 012085.