The Cold Radon Emanation Facility

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Abstract. Radon emanation will continue to be an important background in the next generation of rare-event searches. To aid radon-background mitigation and to improve understanding of its emanation, a dedicated Cold Radon Emanation Facility (CREF) has been developed. The facility is housed at the Rutherford Appleton Laboratory (RAL) in the UK and aims to deliver a sensitivity < 0.1 mBq to ^{222}Rn emanated from materials into emanation chambers at temperatures of relevance to liquefied Xe and Ar rare-event searches. As the name suggests, the CREF vessels can be cooled down to 77 K for measurements of the emanation rates of a sample as a function of temperature. A Si-PIN-diode detector developed by the University of Tokyo is used to detect α-particles resulting from the decay of ^{222}Rn. This report outlines the facility and presents the first detector efficiency, background and minimum detectable activity results.

THE COLD RADON EMANATION FACILITY OVERVIEW

The Cold Radon Emanation Facility is a dedicated cryogenic radon emanation system. The facility can be operated with 2 emanation chambers. The first is a small, 2.7-L chamber capable of operating at several fixed temperatures. The second is a large, 200-L chamber that can be cooled and stabilised at temperatures down to 77 K. The large chamber allows measurements of large material samples to be conducted whilst establishing their emanation rate as a function of temperature. The sensitivity that will be achieved through this second mode of operation is subject to the R&D to be performed, and is dependent on factors including the background from the chamber and the abilities to release emanated radon from the cooled chamber, concentrate the sample within the CREF concentration line, and finally transfer it to the facility's dedicated radon detector. However, performing measurements to profile the temperature dependence of radon emanation from materials does not necessarily depend on achieving sub-mBq sensitivities, since high-rate materials may be used to ensure sufficient signal-to-noise. Nonetheless, the CREF R&D aims to maximise sensitivity through all modes of operations such that, in addition to low-temperature high-sensitivity assays of small samples, large materials with low intrinsic radioactivity required for the construction of next-generation experiments may also be characterised directly. The R&D to be performed with CREF includes the investigation of transfer of radon from cooled samples within chambers through to the detector volumes, likely attachment of radon emanated from samples onto chamber surfaces, and the use of gases and liquids surrounding the samples to maximise collection and transfer of radon for measurement. The large chamber can of course be used for room-temperature measurements without operations of its surrounding cryogenics. The facility will also be used to test radon barriers and material surface treatments that can effectively inhibit the release of radon emanation. Demonstration of the efficacy of coatings, epoxies, and other treatments on the suppression of radon emanation from specific materials at low temperatures would alleviate some of the constraints on material selection, presenting opportunities for innovation in future experiments.

Radon Concentration Line

Due to the sensitivity requirement of CREF (< 0.1 mBq), the facility utilises a radon concentration line (RnCL). The RnCL was developed for the SuperNEMO project, and enables the passing of large volumes of carrier gas through an emanation chamber, collection of its radon content, and its transfer to an electrostatic detector [1, 2]. To minimise backgrounds the pipework is stainless steel, and the valves are metallic. The nitrogen carrier gas is initially directed through 0.5-micron particle filters. It is then flowed through a radon removal system in the form of a cylindrical carbon scrubber filled with activated charcoal, kept inside an ultra-low freezer (Thermo Scientific TLE30086V), held at 193 K. Once cleaned, the carrier gas is passed through the emanation chamber and then towards a cold radon trap filled with high purity active charcoal (CarboAct High-Purity Activated Charcoal, 0.4–2.0 mm). The trap is made from stainless steel, and utilises zirconiated welding. The trap is heated to release the absorbed radon to the detector.

Electrostatic Detector and DAQ

The electrostatic detector has been developed at the University of Tokyo (UoT) [3]. The detector incorporates an 80-L stainless steel vessel which has been electropolished to a roughness of < 0.8 μm. An electric field can be applied across this vessel when it contains a radon sample mixed with carrier gas, causing positively charged radon daughters such as ^{218}Po and ^{214}Po to be attracted towards the PIN photodiode (HAMAMATSU-S3204-09). These isotopes are of interest due to their large α-energy separation, high collection efficiency and high ion fractions. As these species decay, the energies of their ejected α-particles are measured through their energy deposits on the photodiode. The rate of radon emanation from a source material is reconstructed from these measurements by correcting for the detector and transfer efficiencies measured during dedicated runs with radon sources of known activity.

All detector electronics including a ceramic feedthrough for the photodiode are housed inside the Faraday-cage lid of the detector, as visible in Figure 1. The photodiode is windowless to minimise the energy loss of striking α-particles; it is bare to minimise backgrounds. Connected to the diode are the amplifier circuitry and a high-voltage (HV) divider. A voltage of -1900 V is passed through this divider, and separated into -100 V, which is used as the p-layer bias voltage of the diode, and -1800 V, which is supplied to the diode face and generates an electric field inside the chamber, with the vessel being held at ground. Deposited charges from α-particles are converted to voltage pulses by a pre-amplifier with a DC power supply of $\pm$ 12 V. These signals are passed to an ADC board manufactured by the UoT [4]. The amplitude of the signal is converted into a ADC channel number by a peak-holder circuit and the 8-bit ADC, and the output is sent to a Raspberry Pi. The ADC board is currently programmed such that data is collected every 10 minutes, but the minimum time interval that can be achieved is 2 seconds.

FIGURE 1. Image of the 80-L CREF electrostatic detector.

Emanation Chambers and Modes of Operation

The CREF system has the ability to measure the radon activity of a sample through either a 1- or 2-step transfer mechanism. The method used depends on the size of the material being assayed and the amount of nitrogen carrier gas required. It is preferable to use helium rather than nitrogen as a carrier, as electropositive impurities in nitrogen can neutralise ions, negatively impacting the detection efficiency. However tests undertaken at UCL on the CREF detector have confirmed the inability to use helium as a carrier, due to a large current draw at low bias voltage.

The first mode of operation will measure the radon emanation from a sample using the 2.6-L chamber. This is analogous to operations performed at UCL during the LZ cleanliness campaign [5]. The samples will be enclosed inside of the vessel for a time that allows the radon concentration to approach equilibrium. The emanated radon will then be transferred to the detector, bypassing the radon trap, due to the small volume of the chamber in comparison with the detector vessel. In the second mode of operation, the small chamber will be cooled to enable high-sensitivity single-temperature emanation measurements. The small emanation chamber may be cooled using an immersion cooler (capable of cooling to 183.15 K), or through direct immersion into a bath.

In the final mode of operation, a sample will be introduced into the large, 200-L chamber, and the surrounding cryogenic system will be used to cool the vessel as the sample emanates. The 500-L cryogenic housing of CREF

consists of an inner and outer thermal shield which are separate from the emanation chamber. Gaseous nitrogen from dewars is passed into the outer shield and circulated inwards. Eventually, LN can accumulate in the inner shield through this process. The emanation chamber is in both direct and indirect thermal contact with these shields. As the vessel's temperature is decreased, radon will be collected from the chamber at various times. Due to the size of this gas volume, the collected radon will be passed into a radon trap before being transferred into the detector.

DETECTOR EFFICIENCY, BACKGROUND, AND MINIMUM DETECTABLE ACTIVITY

To measure the detector efficiency, the activity of a calibrated source was measured. This source is a 1.32-kBq flow-through ^{226}Ra source (Pylon Electronics, RN-1025) which can be implemented into the RnCL pipework. Before this measurement was conducted, residual activity inside the source was cleared by flushing it with carrier gas. The source valves were then closed to allow an activity build up for time T. The source was then opened, and the radon content transferred to the detector. The activity of radon, $A\left(^{222}\mathrm{Rn}\right)$ that has built up in a time T is given by

$$A\left(^{222}\mathrm{Rn}\right) = A\left(^{226}\mathrm{Ra}\right)\left(1-e^{-T\lambda}\right), \tag{1}$$

where $A(^{226}\mathrm{Ra})$ is the source activity, and λ is the decay constant of ^{222}Rn. A correction is applied to Equation 1 to account for the increase in activity over the time in which the radon is transferred to the detector. With this factor taken into account, a source activity of $A(^{222}\mathrm{Rn}) = 12.62 \pm 0.50$ Bq was found to have accumulated. To extract the detection efficiency of ^{218}Po and ^{214}Po, the energy window (ADC window) corresponding to each α-peak is selected, and the number of decays in this window plotted against time, as shown in the left panel of Figure 2. To determine the initial activity of both isotopes, each population is fitted with an iterative decay equation, the order of which corresponds to its level down the chain with respect to ^{222}Rn (with ^{222}Rn being zeroth order). The extracted efficiencies are $30.4 \pm 1.2\%$ and $41.1 \pm 1.6\%$ for ^{218}Po and ^{214}Po respectively. Both of these efficiencies surpass those achieved using the previous UCL detector [1, 2], due to the improved detector characteristics, such as its vessel geometry, and the choice of windowless diode. The dominant uncertainty in the resulting efficiencies comes from the quoted activity of the source used. The efficiency of ^{218}Po is lower than ^{214}Po, as expected from previous studies at UCL [1, 2]. It should be noted that the maximum possible efficiency of this type of detector is $\sim 50\%$, as only half of the α-particles ejected will hit the photodiode.

Our initial investigations have focused on the sensitivity of the CREF detector. A 24-day background measurement resulted in a detected ^{214}Po event rate of 1.38 ± 0.24 counts/day, implying the internal emanation of the detector is 3.36 $\pm$ 0.59 atoms/day. The detector efficiency for ^{214}Po and its emanation rate can be used to find the Minimum Detectable Activity (MDA) of the CREF detector. The MDA referred to in this section is defined in *Radiation Detection and Measurement* by G. F. Knoll [6] and represents the detection-system sensitivity at a 90% confidence level (CL). If a sample with activity A_S (A_S/λ radon atoms) is introduced into the detector with an intrinsic background of A_D, the number of radon atoms in the detector at a given time can be found by using

$$\frac{dN}{dt} = -\lambda N + A_D. \tag{2}$$

In addition we assume that before a measurement the detector is flushed so $N = A_S/\lambda$ at $t = 0$ can be applied as a boundary condition when integrating Equation 2, giving $C = A_S/\lambda - A_D/\lambda$ and

$$N = \frac{A_D}{\lambda}\left(1-e^{-\lambda t}\right) + \frac{A_S}{\lambda}\left(e^{-\lambda t}\right). \tag{3}$$

This is the sum of the background term and the exponentially decaying signal term [2]. The number of signal events, S, expected to have been detected after a given time T, using a detector with detection efficiency ε, with a detector background of zero is

$$S = \varepsilon\int_0^T \lambda N dt = \varepsilon\int_0^T A_S e^{-\lambda t} dt = \varepsilon\frac{A_S}{\lambda}\left(1-e^{-\lambda T}\right). \tag{4}$$

Similarly to find the number of expected background events B, the signal activity is set to zero so that

$$B = \varepsilon\int_0^T \lambda N dt = \varepsilon\int_0^T A_D\left(1-e^{-\lambda t}\right) dt = \varepsilon A_D T - \frac{\varepsilon A_D}{\lambda}\left(1-e^{-\lambda T}\right). \tag{5}$$

The efficiency of the CREF detector has been measured as $\varepsilon \approx 41.1\%$, and the internal emanation results in a detected rate of ≈ 1.38 cpd, equivalent to a detector activity of $A_D \approx 0.039$ mBq. The MDA for the detector can be calculated by first finding B using Equation 5. The minimum number of signal events detectable for a given CL from a normal approximation (in the low-count Possion regime) is given by

$$S_0 = 2E(E + \sqrt{2B}), \tag{6}$$

where $E = \text{erf}^{-1}(2\text{CL} - 1)$ [1, 2]. The MDA is identified as A_S when $S = S_0$ in Equation 4.

Figure 2 shows the resulting detector MDA versus time, T. This plot shows that the CREF goal of < 0.1 mBq screening capabilities is achievable with this detector. For comparsion, the MDA of the previous UCL detector has been included to highlight the how the improvements made by the UoT on the electrostatic detector have impacted MDA [1, 2].

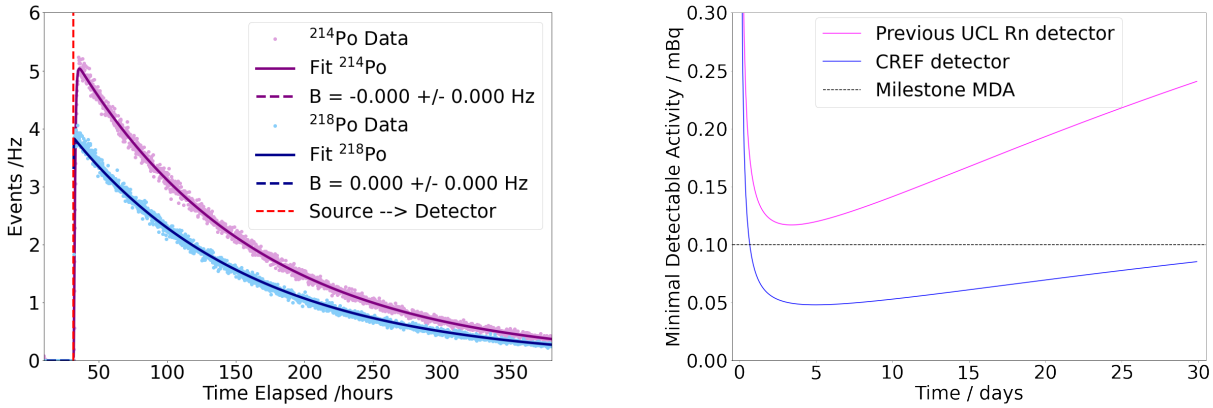

FIGURE 2. Results of the first CREF detector efficiency measurement. **Left:** Detected event rate of ^{214}Po and ^{218}Po when 12.6 Bq of ^{222}Rn is introduced into the detector volume. The resulting peak event rate of ^{214}Po and ^{218}Po is 5.183 $\pm$ 0.072 Hz and 3.843 $\pm$ 0.076 Hz respectively. **Right:** The MDA for the CREF detector over the duration of a measurement.

CONCLUSION

This report has provided a summary of the CREF facility, as well as presenting the first detector efficiency, background and minimum detectable activity results. The conclusion of these results are that the CREF goal of < 0.1 mBq screening capabilities may be achievable with this detector, if transfer efficiencies are sufficiently high and other backgrounds are sufficiently low. The facility is currently in a characterisation phase, with the first cold emanation measurement on track to take place in the summer of 2022.

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