Chapter 1

Radon Emanation Measurements

The radon detector and the RnCL have both demonstrated excellent performance and, when used in conjunction, the capability to reach and surpass the target radon sensitivity required for the SuperNEMO demonstrator. All the radon measurements are carried out for two main purposes: screening to select the cleanest possible material; and building up the background model based on measurements obtained from individual components. The RnCL allows the monitoring of radon emanation from quarter trackers during and after construction providing essential information on meeting the radiopurity requirement. The ability to measure a fully instrumented tracker volume is a key aim as radon emanation is area and geometry dependent. Some key measurements carried out using the radon detector alone, and in combination with the RnCL, are detailed here, including the measurement of three demonstrator quarter trackers, and radon emanation from the SuperNEMO gas system. The radon emanation measurement facility is also used to measure samples for the LZ experiment.

1.1 Sample Measurements

Having established the background levels of the emanation chamber in Section **??**, the chamber was then used for sample emanation tests of SuperNEMO and LZ detector components and construction materials.

1.1.1 Feedthrough Emanation Measurements

Feedthroughs are used to deliver HV to the tracker cells and connect the anodes and cathodes of the cells to the tracker front-end electronics. Each feedthrough consists of a stainless steel frame, and CuBe pins held by the injected Duracon. There are 170 feedthroughs in the Demonstrator's tracker, and 28 of them were used as a sample to test the radon emanation level. The feedthrough sample was cleaned using IPA in the cleanroom laboratory and then inserted into the emanation chamber. The feedthroughs were arranged loosely to make sure all the surfaces can be easily flushed (see Figure 1.1). The flange was then sealed using a new copper ring and flushed with helium at 20 lpm for 15 minutes — 300 litres of helium in total, which is more than 100 volumes replacement of the 2.6-litre chamber. After the flush, the chamber was sealed under atmospheric pressure and left for emanation.

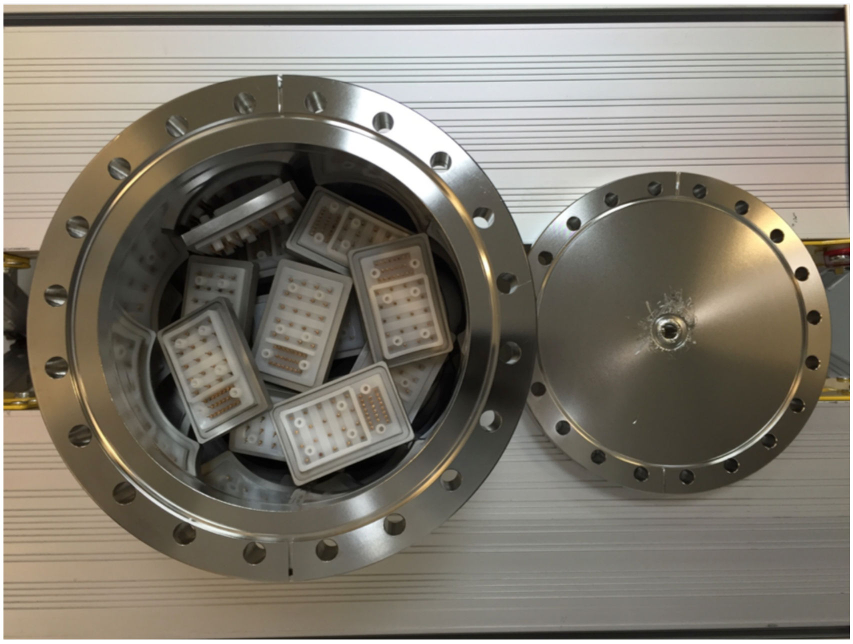

Figure 1.1: The insertion of feedthroughs into the emanation chamber

Before starting the new background measurement, the detector was flushed with clean helium to remove any residual radon. Then, after 10 days, the gas inside the chamber was flushed into the detector for measurement. Due to the low activity of emanated radon, a period of at least one week was required to gather statistics in order to determine the radon emanation level of the feedthrough as shown in

Figure 1.2.

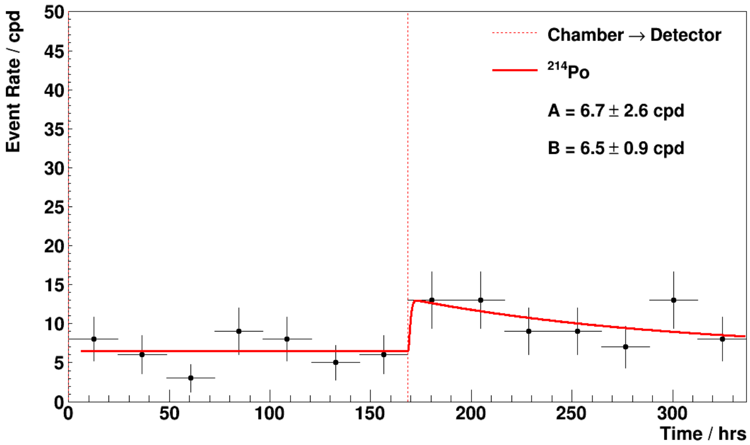

Figure 1.2: Event rates of ^{214}Po during the first feedthrough radon emanation measurement

Due to the low activities of the samples, there are always fluctuations observed during a measurement. Also due to the radon harbouring effect as explained in Chapter **??**, a second measurement should always be carried out. The emanation chamber

is flushed again with 300 litres of helium and sealed under atmospheric pressure for emanation over another two weeks. Then a second transfer was made for the measurement following the similar procedures as the first measurement. The second measurement shows a result consistent with the first one (see Figure 1.3).

1.1.2 A List of Sample Emanation Measurements

To measure the radon activity of the SuperNEMO detector during the construction, all materials and detector components are screened for their radon emanation level. The UCL electrostatic radon detector has also been used for measuring samples for the LZ experiment. The final results of the sample measurements carried out by the author are listed in Table 1.1.

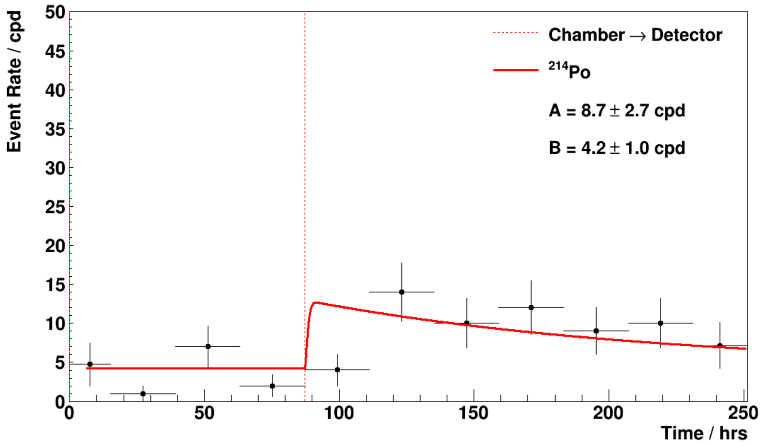

Figure 1.3: Event rates of ^{214}Po during the second feedthrough radon emanation measurement

Experiment	Sample	Radon emanation
SuperNEMO	Feedthrough	0.18 ± 0.06 (mBq/kg)
SuperNEMO	Black Mumba (RTV strips)	0.10 ± 0.08 (mBq/kg)
SuperNEMO	Protective Mylar	0.75 ± 1.20 (mBq/m^3)
LZ	Derlin Disks	5.6 ± 1.8 (μmBq/each)
LZ	O-rings	15.2 ± 1.4 (μBq/each)
LZ	PMT base	5.0 ± 0.9 (μBq/each)
LZ	resistors	17.9 ± 5.7 (μ/g)

Table 1.1: The results of radon emanation measurements from detector components and construction materials of SuperNEMO and the LZ experiment, all measured by the electrostatic detector.

1.2 Gas System measurement

The tracker gas of SuperNEMO is made up of 95% helium, 1% argon and 4% ethanol. To obtain the accurate proportions, a dedicated gas supply system has been developed at UCL. The ^{222}Rn emanation level of the SuperNEMO gas system has been reduced, comparing to the NEMO-3 gas system. It is compact and transportable such that it can be used both for the commissioning of the tracker and for running the experiment in its final configuration at LSM.

The biggest part of the gas system is a 50-litre cylinder bubbler made of stainless steel for better radiopurity. As a carbon radon trap similar to the RnCL trap to filter the output from the gas system cannot be installed, as it would result in the removal of ethanol as well, the gas system is instead connected directly to the tracker. The volume of the gas system is relatively small, so that at the proposed flushing rate (1-2 m^3/h), all ^{222}Rn emanated from the gas system components or diffused into the gas will be flushed into the tracker volume within the half-life of ^{222}Rn. Hence, it is strictly required that radon emanation from the gas system is < 0.2 mBq, corresponding to 10% of the total tracker radon budget 2 mBq.

In order to verify the radon emanation level of the Gas System — three different measurements, spike measurement, flowthrough measurement and the RnCL measurement — were carried out.

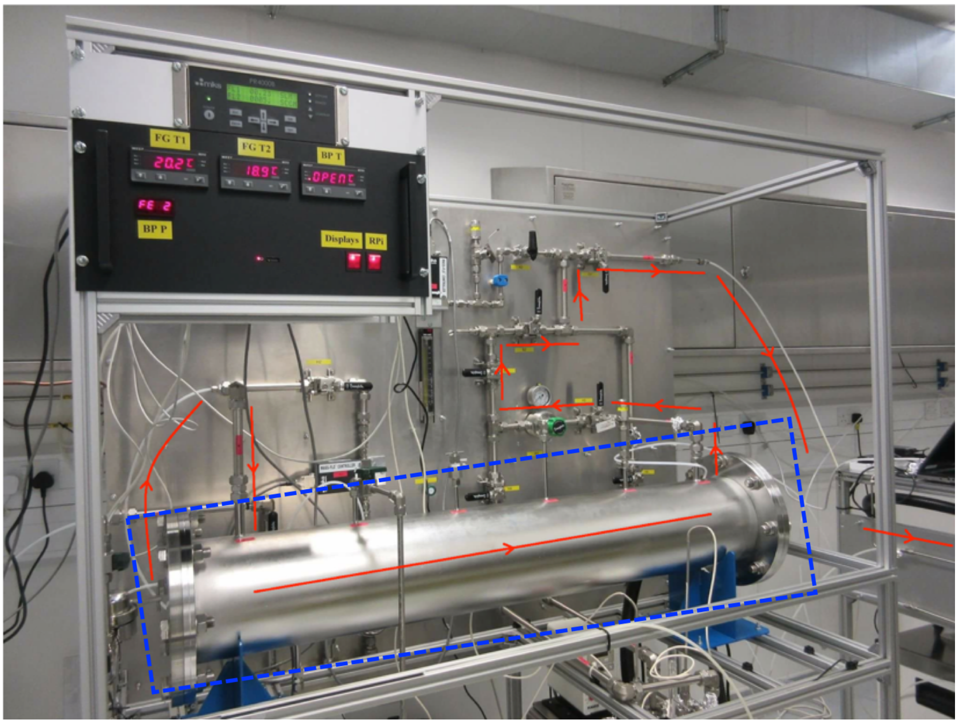

Figure 1.4: The gas system in reallife. The primary region for the radon emanation test is labelled in red [1], and the chamber in blue sashed box is the main bubbler.

1.2.1 Removing Ethanol

Ethanol can affect radon emanation and can also block the active carbon trap of the RnCL. As such, prior to carrying out the radon emanation measurement of the gas system, it is essential that all the ethanol inside the gas system is removed. The requirement of the residual ethanol level for carrying out the RnCL measurement is stringent, < 10 ppm, hence a residual gas analyser (RGA) was used to monitor the content of the output gas from the gas system. The main bubbler was drained and then flushed with nitrogen and helium to remove ethanol [2].

1.2.2 Flowthrough Measurement

The primary purpose of the flowthrough measurement is a quick check to ensure that there is no major emanation, but required sensitivity cannot be reached with this method. In this measurement, the gas system was set up to flow at 4.2 lpm rather than mimicking the real operation flowrate 14 lpm, taking advantage of the low flowrate to reach better sensitivity.

The procedures are as below:
1. Nitrogen was first used to flush the gas system for 24 hours prior to starting the measurement.
2. Use 500 litres of zero grade helium (10 volumes of the gas system) purging through the gas system to replace the nitrogen before starting the measurement for better detection efficiency.
3. Helium was flown from the gas system directly through the detector to exhaust at 4.2 lpm over 3 days.

Results of the flowthrough method measurement are shown in Figure 1.5

No increase of radon level activity was observed in this measurement; thus, only a limit could be extracted. Since the measurement was carried out while flushing, the flow suppression factor has been taken into account.

$$A_D = \frac{A_0}{1+\frac{\phi}{\lambda V}} + A_B \tag{1.1}$$

where A_D is the observed activity;
A_0 is the true activity;

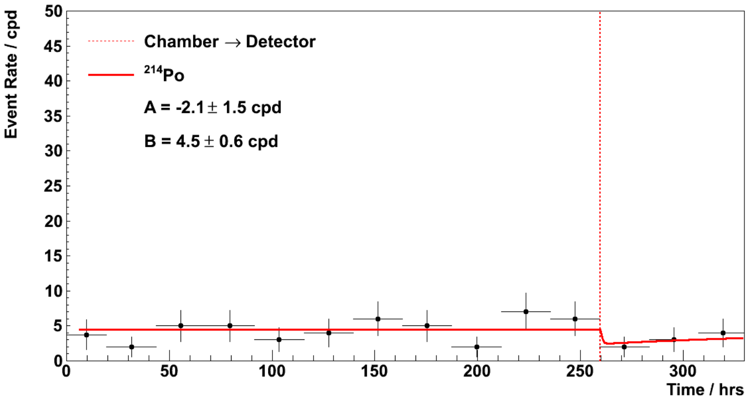

Figure 1.5: Event rates of ^{214}Po during the radon emanation measurement of the gas system using the flowthrough method [2].

A_B is the background activity of the detector;

λ is the decay constant of ^{222}Rn;

$1+\frac{\phi}{\lambda V}$ is the suppression factor.

Taking the flowrate ϕ of 4.2 lpm and a volume, V, of 120 litres(the 50-litre gas system and the 70-litre radon detector), the suppression factor is calculated to be 280. From the uncertainty of the observed ^{214}Po activity, a limit can be placed at $<$ 25 mBq at 90% CL. The quick flowthrough measurement confirmed that no significant radon emanation was observed.

1.2.3 Spike Measurement

The gas system can be seen as a big emanation chamber so that a spike measurement of the gas system is similar to the measurements of samples in the emanation chamber. However, it is slightly different from the sample emanation measurement due to the large volume of the bubbler. The detector is calibrated at 1.35 bar, and also it is not certified above 2 bar in term of sealing. To avoid too much overpressure inside the detector, only 25 litres of gas will be transferred into the detector as usual, which means the sample volume is smaller than the volume of the gas system.

Procedures of the measurement are as below:

1. Flush the gas system with helium from the J-trap at 20 lpm for 30 mins, totally purging 12 volumes of gas as replacements and then seal it under atmospheric pressure. In the meantime, a leaking test is carried out using a Helium sniffer. The gas system is then left for radon build-up over 17 days to reach equilibrium.
2. Flush the gas line and clear the radon detector to start the background measurement.
3. After the emanation period, check the detector background to ensure it is low and stable and flush the entire gas line except the gas system to remove residual radon in the gas line and then immediately transfer 25 litres of helium from the gas system into the radon detector.

Results from the spike measurement are shown as in Figure 1.6.

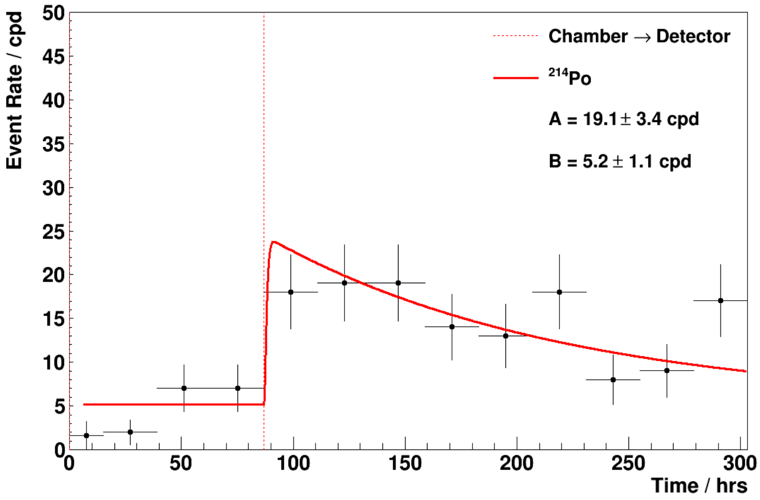

Figure 1.6: Event rates of ^{214}Po and ^{218}Po during the spike method radon emanation measurement of the gas system.

It should be noted that the transfer efficiency and the proportion of the volume should be counted in when extracting the activity. The transfer efficiency, ε_{trans}, was previously measured to be $70.1 \pm 0.9\%$ [3], and the proportion of the volume transferred is 50% (25 litres of helium transferred V_{trans} over the 50-litre gas system V_G). From the ^{214}Po result, the activity can be extracted as 1.97 ± 0.31 mBq using

Equation 1.2.

$$A_C = \frac{A\ (cpd) \times 1000\ (mBq)}{\varepsilon_D \times (\varepsilon_{trans} \times \frac{V_{trans}}{V_G}) \times (1 - e^{-\lambda t}) \times 86400\ (secs/day)} \tag{1.2}$$

1.2.4 RnCL Measurement

The RnCL measurement of the gas system is similar to the C-Section measurement as seen in Section 1.3, and the schematic of the measurement setup is shown in Figure 1.7. The volume of the gas system is dominated by the large bubbler, which has comparable size to the 70-litre electrostatic detector. The trapping transfer efficiency (Section **??**) can be reliably used if the same flowrate, trap temperature and timing conditions are replicated. and the schematic of the measurement setup is shown in Figure 1.7.

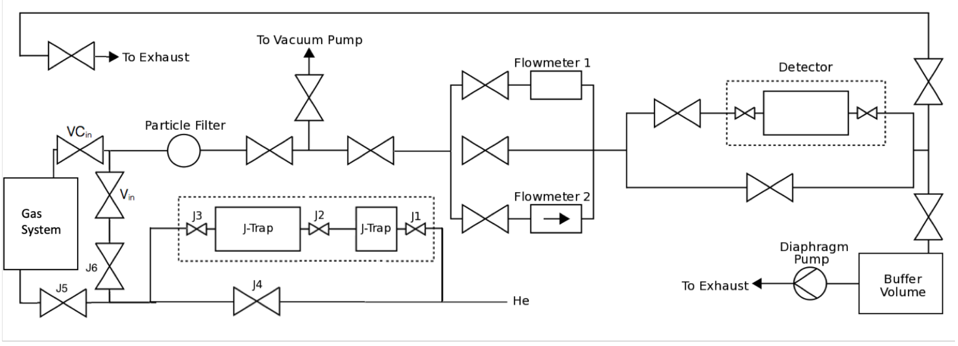

Figure 1.7: The system setup for the RnCL method measurement of the gas system.

the activity of radon introduced right after the transfer is [4]

$$A_D = \varepsilon_{tr} A_C (1 - e^{-\lambda T_C}) + \frac{\varepsilon_{tr}\varepsilon_T(T_f) f a^{eq}_{GS}}{\lambda}(1 - e^{-\lambda T_C})e^{-\lambda T_{trans}} \tag{1.3}$$

where:

λ is the decay constant of ^{222}Rn;

ε_{tr} is the transfer efficiency;

ε_T is the trapping efficiency;

T_C is the time between clearing the trap and detector transfer (1705 min);

T_f is the time that the line is in contact with the trap (1200 min);

T_{trans} is the time between stopping trapping collection and detector transfer (240 min);
A_D is the radon activity in the electrostatic detector;
A_C is the radon activity of the carbon trap;
A_{GS}^{eq} is the equilibrium activity inside the gas system, given by [4]

$$a_{GS}^{eq} = \frac{A_{GS} + f_{in} a_G / \lambda}{V_{GS} + f_{in} / \lambda} \tag{1.4}$$

f_{in} is the input flowrate of gas;
A_{GS} is the radon activity of the gas system;
V_{GS} is the volume of the gas system.
The carrier gas radioactivity a_G are suppressed by the J-Trap. The measurement details of a_G were described in [5].

In the most recent calibration, the results of the transfer efficiency is 92.3% and the trapping transfer efficiency is 73.58%. The radon activity of the carbon trap, A_C, was measured as 0.48 ± 0.14 mBq (based on ^{214}Po results) with a carrier gas radioactivity a_G of 20.2 ± 12.5 μBq/m^3 [5]. The radioactivity of the gas system can be calculated as 1.21 ± 0.38 mBq.

$$A_{GS} = (1 + \frac{f_{in}}{\lambda V_{GS}}) V_{GS} a_{GS}^{eq} - \frac{f_{in} a_G}{\lambda} \tag{1.5}$$

The result from the RnCL measurement and result from the spike method are consistent at 1.5 sigma level. Moreover, all results measured by three different methods have confirmed that the radon emanation of the gas system is < 2 mBq, considering the flow suppression factor 9.71 (at 1 m^3/h, see flow suppression), this number can be further reduced to < 0.2 mBq, which takes $\sim$ 10% of the demonstrator radon budget.

1.3 Quarter Tracker (the C-Section) Measurements

The radon radiopurity budget for the SuperNEMO Demonstrator Tracker gas is $<$ 0.15 mBq/m^3. The sensitivity of the RnCL is calibrated at 8.4 m^3 is $<$ 20 μBq (see Section **??**), which is more than enough to meet the requirement of measuring the radon emanation level of the SuperNEMO quarter tracker Module (also called C-Section due to its C shape). All C-Sections were built and then sealed for radon emanation test at MSSL before they were eventually sent to LSM. Each tracker contains a stainless steel frame, 504 octagonal drift cells and 48 5-inch PMTs of which 32 are on the x-walls, and the other 16 are gamma vetos. Figure 1.8 shows the drift cell cassettes insertion complected for a C-Section.

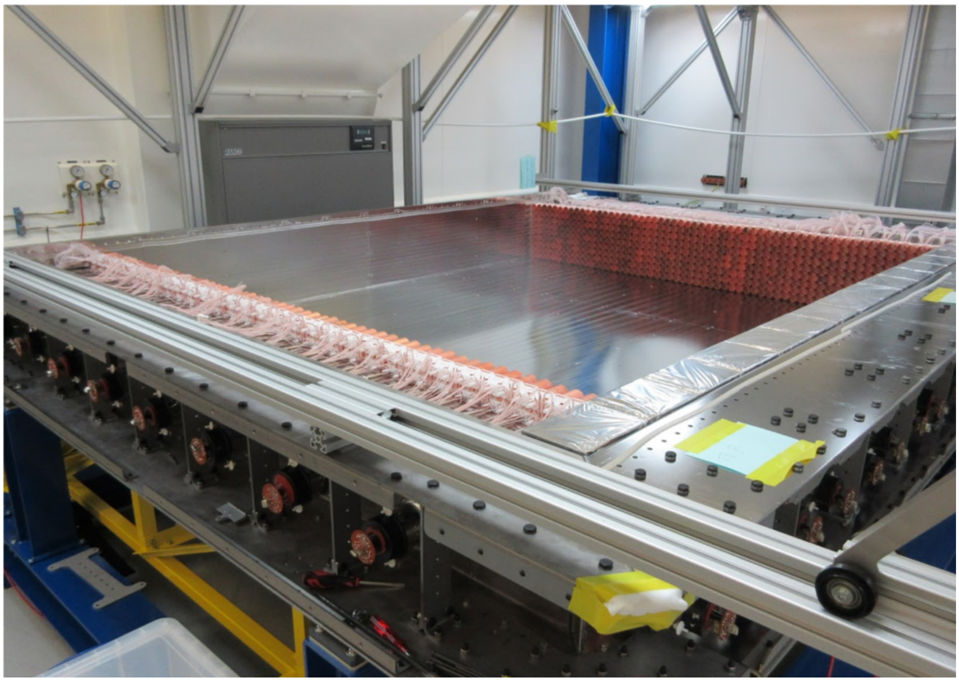

Figure 1.8: A C-Section (quarter tracker) under construction in the cleanroom lab at MSSL.

The main purpose of doing the radon emanation measurements of C-Sections is to obtain the activity from the tracker to build up the background model. The measured activity of the whole tracker is used in the simulation and analysis in Chapter **??**. In addition, the first two C-Sections, C0 and C1, were not only measured after insertion was completed, but also tested before the cassette insertion and halfway during the insertion, to help to monitor the radon level and to point out any positive radon contribution from components so that the contaminated parts can be removed. C-Sections were temporarily sealed with customised gas-sealing plates for the radon emanation test, tracker cell commissioning and transportation, prior to the final full demonstrator assembly when the all four C-Section were joined together with the source frame and calorimeter walls.

1.3.1 Measurement Starting Point

Ideally, the C-Section should be sealed perfectly and left for radon to emanate before the measurement; however, there were always small leaks at this stage. To prevent radon diffusion from the outside, a constant small overpressure supplied by a constant rate of gas flow must be kept inside the C-Section chamber. The C-Section was exposed to the cleanroom air during construction; thus, the components are expected to house some radon as indicated by the radon harbouring hypothesis. As part of the preparation for the measurement, the sealed C-Section must be continuously flushed at 3 lpm using cylindered nitrogen over 18 days to remove residual radon [2]. In addition, the gas purging offered a 2 mbar overpressure inside the C-Section to prevent radon diffusion. Before starting a measurement, the flowrate was increased from 3 lpm to 14 lpm, which can maximise gas flow through the RnCL while keeping a secure overpressure inside the C-Section. Considering no external radon entering the C-Section, the number of radon atoms inside C-Section, N_T can be described by a model:

$$\frac{dN_T}{dt} = A_T + A_G - \lambda N_T - \frac{f_{in} N_T}{V_T} + \frac{f_{in} a_G}{\lambda} \tag{1.6}$$

where

A_T is the intrinsic activity of the C-Section;

A_G is the intrinsic activity of the gas supply line;

λ is the decay constant of ^{222}Rn;

$f_{in}$is the flowrate of input gas;

V_T is the volume of a C-Section which is 3.8 m^3;

a_G is the activity of gas per unit volume from a specific cylinder or the J-trap, depending on the source of the gas.

So the activity of the output gas after the flushing time, t, can be modelled as:

$$A(t) = \frac{A_G + A_T + f_{in} a_G/\lambda}{\lambda'_T/\lambda}(1 - e^{\lambda'_T t}) + A_0 e^{\lambda'_T t} \tag{1.7}$$

where A_0 is the measured activity, and λ' is the modified decay constant defined by:

$$\lambda'_T = \lambda + \frac{f_{in}}{V_T} \tag{1.8}$$

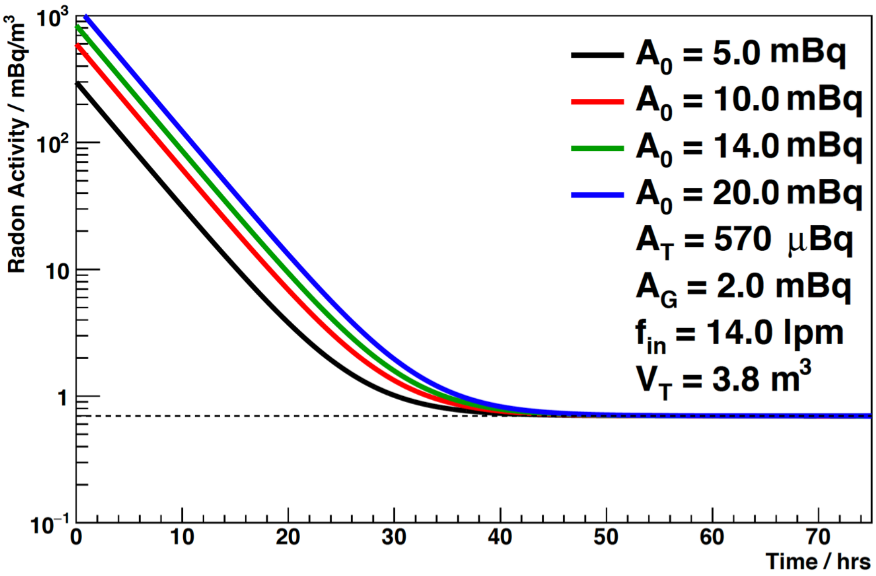

Figure 1.9: Activity inside the C-Section whilst flushing at 14 lpm prior to the radon measurement.

The measurement should only start after the output activity from C-Section reaches an equilibrium. Thus prior to the measurement, the C-Section was flushed for at least 50 hours, see Figure and during this stage, the output flowrate was only 7 lpm due to leaks.

1.3.2 Anti-radon Tent

It has been observed that the radon level of the cleanroom air fluctuated from 2-3 Bq/m^3 to 20-30 Bq/m^3, which is not low and stable enough and as such it is necessary to isolate the C-Section inside an anti-radon tent under a constant gas-flow to provide a cleaner environment. Double-layer transparent polyethene sheets were heat sealed together and then taped onto the ground to build the anti-radon tent. There are four flushing points installed at equal intervals on the tent and purged at 60 lpm to provide a small overpressure in the tent to prevent radon diffusion [6]. A RAD7 detector was installed to monitor the background level of radon in the anti-radon tent during the C-Section measurement. The result, ~ 0.1 Bq/m^3, shows a reduction of 2 orders of magnitude, indicating the anti-radon tent successfully reduced radon from outside.

Figure 1.10: C2 in the annti-radon tent during the measurement.

1.3.3 Procedures of the C-Section Measurement

During the 50 hours' flushing RnCL was connected to the C-Section using 6 mm nylon pipe which has been tested of no positive radon emanation contribution. A diaphragm pump was installed at the exhaust of the RnCL to promote gas purging because the low overpressure inside the C-Section chamber is not enough to maintain the required flowrate.

After 50 hours of flushing, the measurement can be started. Firstly, the RnCL trap was heated to release residual radon and flush radon away. Once the trap was cleaned, it was then sealed under atmospheric pressure, and the time zero of t_C is set. The trap was then cooled down to prepare for the radon trapping stage. When the temperature reached - 40°C, start trapping by diverting nitrogen from the C-Section through the trap, at 7 lpm, and out to exhaust. This trapping stage lasts for 20 hours. After trapping, the trap was sealed again under atmospheric pressure, left to warm up to room temperature and then heated to release the absorbed radon. Upon reaching 220°C, transfer radon from the trap into the electrostatic radon detector by purging 25 litres of helium through the trap.

1.3.4 C-Section Activity Extracting

As mentioned, there were leaks in the C-Section, through which radon can be lost. To extract the real intrinsic activity of the C-Section, it is necessary to model the observed radon. Prior to the start of the measurement, the C-Section is flushed at 14 lpm for 50 hrs to reach equilibrium. The activity inside the C-Section a_T^{eq} can be described by [4]:

$$a_T^{eq} = \frac{A_T + A_G + f_{in} a_G/\lambda}{V_T + f_{in}/\lambda} \tag{1.9}$$

such that activity A_T of the C-Section is

$$A_T = (1 + \frac{f_{in}}{\lambda V_T}) V_T a_T^{eq} - A_G - \frac{f_{in} a_G}{\lambda} \tag{1.10}$$

1.3.5 C-Section Measurement Results

During the radon emanation measurements of the first two C-Sections, C0 and C1, the Delrin caps used as the Geiger cell carriers showed an excess contribution of 4

± 1 mBq for each C-Section. These components were replaced for the construction of the C2 and C3. The measurement result of the completed C2 are shown in Figure 1.11.

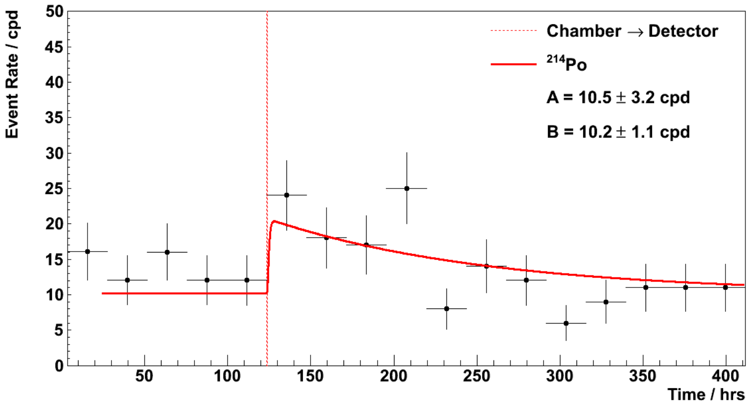

Figure 1.11: Results from the radon emanation measurements of the completed C2 [2].

The a_T^{eq} was calculated to be 0.057± 0.017 mBq based on the ^{214}Po result. And through Equation 1.10, the activity of of C2 was calculated:

$$A_T = 4.36 \pm 1.31 \mathrm{mBq} \tag{1.11}$$

which translates to

$$a_T = 1.15 \pm 0.34 \mathrm{mBq/m^3} \tag{1.12}$$

The results of the first three C-Sections are summarised in Table 1.2. Assuming the

	activity (mBq)
C0	11.37 ± 1.44 mBq
C1	$15.26\ ^{+2.50}_{-4.00}$ mBq
C2	4.36 ± 1.31 mBq

Table 1.2: Summary of C-Section measurement results.

radon emanation level of C3 is an average of the first three C-Sections, the activity of the entire SuperNEMO Demonstrator tracker is:

$$A_D = 41.3 \pm 4.7\text{mBq} \tag{1.13}$$

translate to

$$a_D = 2.7 \pm 0.3\text{mBq} \tag{1.14}$$

This result shows that a radon activity of 0.28 ± 0.07 mBq/m^3 can be achieved for the SuperNEMO Demonstrator by continuously flushing with clean tracker gas at 1 m^3/h, providing a suppression factor of 9.71. This activity number is used to model the the SuperNEMO background in simulation and analysis described in Chapter **??**.

Bibliography

[1] X. R. Liu. Gas System Radon Emanation Measurement. *Internal Note*, DocDB:4161, 2016.

[2] X. R. Liu. *Low background techniques for the SuperNEMO experiment*. PhD thesis, University College London, 2017.

[3] B. Soule. Emanation Setup Status and 5 Inch PMT Measurements. *Internal Note*, DocDB:3270, 2014.

[4] J. E. Mott. *Search for double beta decay of ^{82}Se with the NEMO-3 detector and development of apparatus for low-level radon measurements for the SuperNEMO experiment*. PhD thesis, University College London, 2010.

[5] X. R. Liu. Gas Measurement Using Radon Purification System. *Internal Note*, DocDB:3593, 2014.

[6] X. R. Liu. Gas Measurement Using Radon Purification System. *Internal Note*, DocDB:3593, 2014.