Studies on the Reduction of Radon Plate-out on Copper Using Electric Fields

SMU E-SHIELD PROJECT

Approved by ______________________________

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Studies on the Reduction of Radon Plate-out on Copper Using Electric Fields

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Abstract:

I investigate the ability of electric fields to reduce the capability of radon progenies to stick to copper surfaces. Mitigating radon progeny exposure is important to the community of experiments searching for rare processes, requiring low or no backgrounds. Background interactions from gamma particles, neutrons, and alpha particles can mimic predicted signals of certain dark matter or neutrino-less double beta decay processes. ^{222}Rn is a large contributor of background noise in the experiment when detector material or shielding material is stored for periods of time. To understand how electric fields in storage environments can help reduce the ^{222}Rn plate-out, we propose to try to contaminate copper samples protected by an electric field with radioactive sources and then measure actual exposure levels with the XIA UltraLo 1800 alpha particle counter. The experimental set-up includes a pressure cooker where the copper will be stored and exposed to a source of ^{220}Rn, which will serve as a proxy for ^{222}Rn to shorten the length of experimental trials. The copper will be held by a custom shelving unit made out of 3D-printed plastic, providing an insulating framework when the electric field is applied.

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1 Introduction

Radon is a naturally occurring radioactive gas that is common in the environment. Its progeny, resulting from nuclear decay, can attach ("plate-out") to the surfaces of materials and continue to decay. These materials may then be used in the setup of a sensitive experiment, such as those searching for dark matter particles or for the very rare neutrino-less double beta decay process. The decay products from these progenies produce background levels by gamma particles, neutrons, and alpha particle interactions and can mimic predicted signals of dark matter interactions or neutrino-less double beta decay. Reduction of these sources of background events is crucial to achieve the proposed sensitivity level for next-generation experiments in these research areas.

^{222}Rn is a large contributor of background noise in the experiment when detector material or shielding material is stored for periods of time. A study of the radon decay chain suggests that 88% of the radon decay daughters are positively electrically charged[1]. It is possible, therefore, that storing materials in the presence of an external electric field will mitigate the plate-out of radon progeny. To understand how electric fields in storage environments can help reduce the ^{222}Rn plate-out, we create an environment in which the effect of an external electric field can be isolated from other factors. Copper samples are then placed in the environment and exposed to a radon source. The XIA Ultra-Lo 1800 is used as an alpha particle counter, which is sensitive to the decay of certain radon daughters and subsequent alpha particles emission after contamination, we can assess levels after the exposure. The experimental set-up includes a pressure cooker where the copper is stored and exposed to a source of ^{220}Rn, which serves as a proxy for ^{220}Rn to shorten the lifetime of experimental trials. We use ^{220}Rn as a proxy for ^{222}Rn because we are not licensed to have a ^{222}Rn source at this time. However, ^{220}Rn sources are common and require no special licensing beyond SMU requirements. The short half-lives of ^{220}Rn daughters allow for a quick experimental turn around. A custom-shelving unit made out of 3D-printed plastic serves as an insulating support framework for the copper when the electric field is applied.

2 Motivation

Uranium is a common radioactive element found throughout the Earth's crust and other geological layers[2]. The decay chain of uranium is long, but a primary stage in its decay is the production of radon, another radioactive element that is gaseous at room temperature[1]. Near the end of the decay chain of the ^{222}Rn is ^{210}Pb, which is also radioactive and has a half-life of 22.3 years[3]. It is this isotope that is problematic to sensitive experiments, since it lasts a very long time (much longer than experimentalists can wait for it to decay away) and gives off radiation during its own decay that can create significant background for these experiments[4]. Reduction of these backgrounds is crucial since radon background levels can contribute to reducing the sensitivity of experiments that search for Weakly Interacting Massive Particles (WIMPs).

Radon contamination mitigation is the subject of study by a variety of fields, including the medical community (radon is a leading cause of lung cancer[1]), the semiconductor industry (to prevent contamination in otherwise pure semiconductor wafers[5]), and the physics community. Reduction in radon, therefore, has been a quickly growing field of study in the last few decades. The goal in the physics community is to quantify the reduction in radon plate-out that can be achieved by different techniques. Achieving a significant reduction in radon contamination is crucial to advancing the sensitivity of rare process search experiments.

3 Radon Decay Chain

The ^{238}U decay chain (Fig. 2) shows ^{222}Rn and its subsequent progenies along with their half-lives. ^{238}U is the most common isotope of uranium, contributing 99.284% of natural uranium. The focus of reduction efforts in the physics community is on ^{222}Rn, which lies in the middle of the decay chain, resulting from the decay of ^{226}Ra.

The State of Texas has a licensing procedure for the use of radioactive isotopes in scientific research, as well as for other applications. The SMU license does not currently cover the possession of a sufficient ^{222}Rn source for experiments (obtaining the extension to the license is a work-in-progress in the Physics Department). However, ^{220}Rn is available in common material and its possession and use is covered by the existing license. For the experiments described in this thesis, a ^{220}Rn source is employed. The half-lives of the ^{220}Rn source are significantly shorter than those of the ^{222}Rn chain, thus not a concern for running physics experiments since it quickly decays away. Pre-1990s camping lantern mantles are used as the source of ^{220}Rn since its parent isotope, ^{232}Th, is abundant[6].

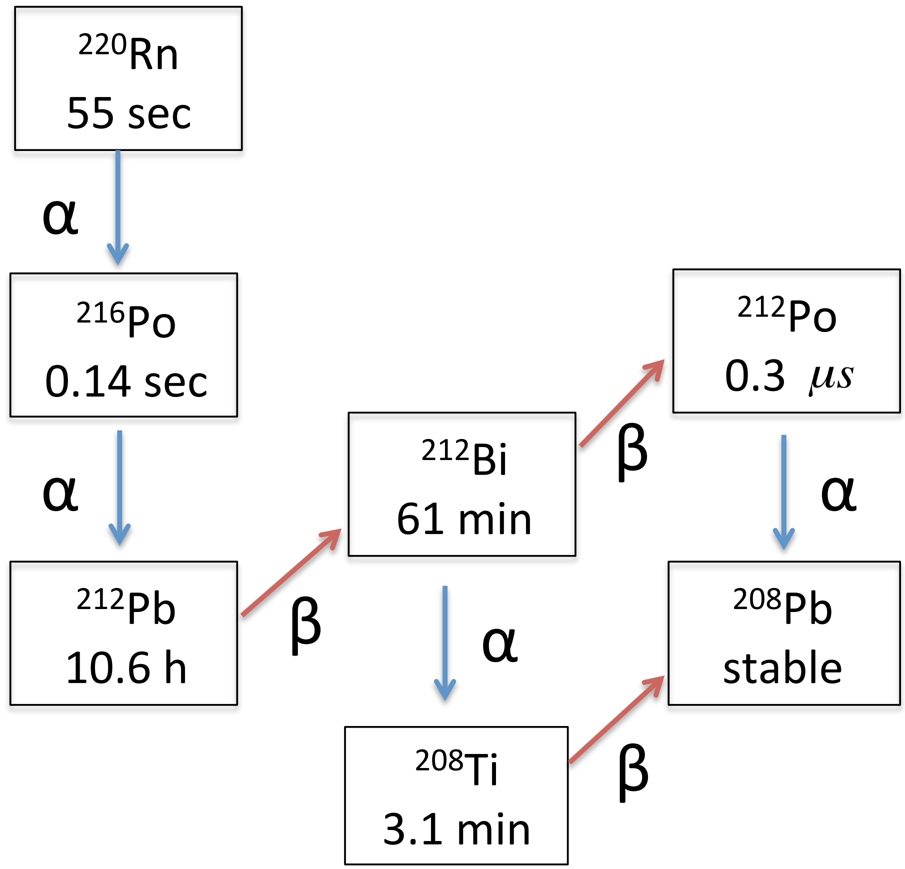

Figure 1 - The ^{238}Thorium Decay Chain Starting at ^{220}Radon

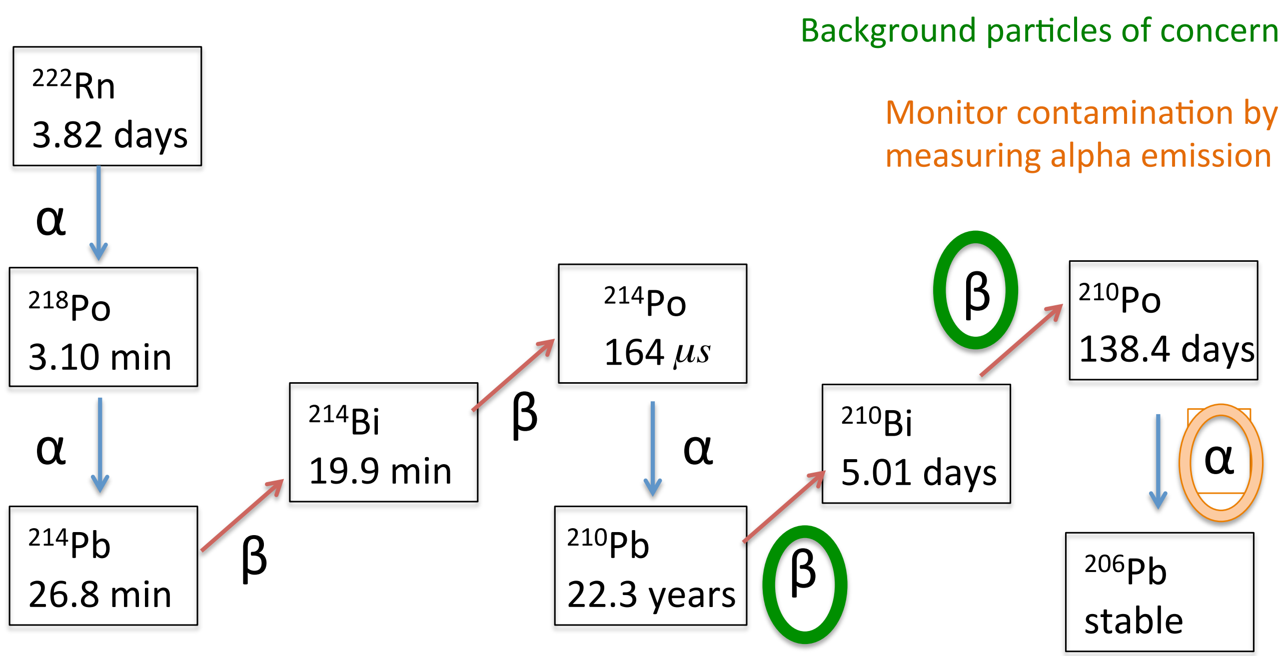

Figure 2 - The ^{238}Uranium Decay Chain Starting at ^{222}Radon

4 Calculation of the Stopping Potential

In this section there are calculations of the electric potential required to stop radon progenies and then reverse their direction of motion under two scenarios. The decay ^{220}Rn ➔ ^{216}Po is used as the example in the calculations. These calculations were performed before the experiment described in this thesis was performed. They were used to motivate the equipment needed for the experiment, and to give a rough estimate of the benefit of using an electric field to stop radon progenies from plating-out on a sample. The stopping potential is calculated for the general setup available for the experiments: a copper source within a pressure cooker with an electric field applied for shielding of the copper. The pressure cooker has a radius of four inches with a height of just over seven inches. The copper samples being exposed are four by four inches and are held by a copper holder of dimensions 4inx4.5inx6in. This allows for a tight setup for the stopping potential to occur. The calculations for the non-thermalized and thermalized ^{216}Po particles provide the minimum and maximum potentials that are needed to be implemented within the design of an electric field to prevent the plate-out of ^{220}Rn and its progenies.

The average kinetic energy of a single radon daughter is calculated using the Boltzmann constant and an assumed room temperature of 295 Kelvin.

$$E_{molecular} = \frac{3}{2} k_B T_{room} = \frac{3}{2}\left(1.38 \times 10^{-23} \frac{J}{K}\right)(295K) = 6.11 \times 10^{-21} J \qquad (1)$$

First, a calculation was made for a daughter particle (^{216}Po) that is thermalized within the pressure cooker. This assumption was made initially to calculate a minimum potential that is needed in the system. First, the average velocity for a thermalized daughter atom was calculated using the isotope mass for the daughter and the average kinetic energy from Eqn. 1:

The average velocity for the thermalized particle is calculated using the molecular energy within the pressure cooker and the mass of the ^{216}Po atom:

$$v_T = \sqrt{\frac{2E_{molecular}}{m_T}} = \sqrt{\frac{2\left(6.11 \times 10^{-21} J\right)}{3.62 \times 10^{-25} kg}} = 184 \frac{m}{s} \qquad (2)$$

Using the velocity found in Eqn. 2, the acceleration required to stop the thermalized daughter isotope within the pressure cooker can be found assuming a 3 cm distance from the radon source to the copper target:

$$a_T = \frac{v_T^2}{2d_{source}} = \frac{\left(184 \frac{m}{s}\right)^2}{2(0.03m)} = 5.64 \times 10^5 \frac{m}{s^2} \qquad (3)$$

Having determined the required acceleration, the electric field can be found using the definition of electric force (F = qE) and Newton's second law (F = ma) just as for the non-thermalized particle:

$$E_T = \frac{m_T a_T}{2e} = \frac{\left(3.62\times10^{-25}kg\right)\left(5.64\times10^{5}\frac{m}{s^2}\right)}{2\left(1.6\times10^{-19}C\right)} = 0.64\frac{N}{C} \quad (4)$$

In order to estimate the potential required to stop this particle, a uniform electric field is assumed (no detailed modeling of a specific electric field configuration was used to refine this calculation). Using this assumption, the potential is calculated as:

$$V_T = E_T d_{source} = \left(0.64\frac{N}{C}\right)\left(0.03m\right) = 0.019V \quad (5)$$

For an estimate of the maximum potential calculation, and to determine the potential that will be used in the set-up, the decay, ^{220}Rn ➔ ^{216}Po+α, needs to be analyzed. This case is where the radon daughter gets the full energy available to it from the decay, which must be stopped by the potential. Natural units are used to simplify the mathematics, thus mass and momentum have the same units of energy. Since the masses of the ^{220}Rn and ^{216}Po particles are very similar, and have been precisely measured to keep all significant decimal places in the calculation:

$$m_\alpha = 3.727379\times10^{9}eV \quad (6)$$

$$m_{Po216} = 2.0307406\times10^{11}eV \quad (7)$$

$$m_{Rn220} = 2.06808060^{11}eV \quad (8)$$

Conservation of momentum for the decay can be used to solve for the momentum of the ^{216}Po particle:

$$m_{Rn220}^2 = m_\alpha^2 + 2\left(\sqrt{m_\alpha^2 + p^2}\sqrt{m_{Po216}^2 + p^2} + p^2\right) + m_{Po216}^2 \tag{9}$$

Solving for momentum using the masses of the particles:

$$p = 2.20249 \times 10^8 \frac{eV}{c} = 1.175440^{-19} \frac{kgm}{s} \tag{10}$$

Using the momentum found in Eqn. 14, the kinetic energy can be easily found by relating momentum and the mass of the ^{216}Po particle. The classical approximation is used for these calculations since the velocity found is much lower than that of the speed of light.

$$KE_{Po216} = \frac{p^2}{2m_{Po216}} = \frac{\left(1.17544 \times 10^{-19} \frac{kgm}{s}\right)^2}{2\left(3.62 \times 10^{-25} kg\right)} = 1.91 \times 10^{-14} J \tag{11}$$

The electric field and potential calculations that were used for the thermalized particles can be repeated to find the worst-case scenario potential needed within the pressure cooker. The velocity of the radon daughter is calculated:

$$v_{NTPo216} = \sqrt{\frac{2KE_{Po216}}{m_{Po216}}} = \sqrt{\frac{2\left(1.9083 \times 10^{-14} J\right)}{3.62 \times 10^{-25} kg}} = 324{,}696 \frac{m}{s} \tag{12}$$

Using the velocity of the ^{216}Po particle to solve for the acceleration assuming a 3 cm distance from the radon source to the copper:

$$a_{NTPo216} = \frac{v_{NTPo216}^2}{2d_{source}} = \frac{\left(324,696\frac{m}{s}\right)^2}{2(0.03m)} = 1.76\times10^{12}\frac{m}{s^2} \quad (13)$$

The electric field needed to stop this is then:

$$E_{NTPo216} = \frac{m_{Po216}a_{NTPo216}}{2e} = \frac{\left(3.62\times10^{-25}kg\right)\left(1.76\times10^{12}\frac{m}{s^2}\right)}{2\left(1.6\times10^{-19}C\right)} = 1.988\times10^{6}\frac{N}{C} \quad (14)$$

Assuming a constant electric field applied throughout the pressure cooker, the potential for the system can be found:

$$V_{NTPo216} = E_{NTPo216}d_{source} = \left(1.988\times10^{6}\frac{N}{C}\right)(0.03m) = 60kV \quad (15)$$

Therefore, the maximum potential, the potential needed to stop a non-thermalized ^{216}Po atom, is approximately 60kV. This potential is much higher than the minimum potential solved by using a thermalized polonium atom particle, so this maximum potential should be taken into account for the design of the system.

The mean free path for the non-thermalized ^{216}Po particle is also of interest since an assumption was made that the distance from the copper to the electric field is short (3 cm). The mean free path should be calculated to determine how far the ^{216}Po atom would travel within the pressure cooker before becoming thermalized in the standard atmosphere in the vessel.

Assuming a standard atmospheric pressure of 100kPa within the pressure cooker:

$$l = \frac{KE_{NTPo216}}{\sqrt{2}\pi d_{Po216atom} P_{room}} = \frac{1.91 \times 10^{-14} J}{\sqrt{2}\pi\left(167 \times 10^{-12} m\right)\left(1.0 \times 10^{5} Pa\right)} = 1.54m \qquad (16)$$

The mean free path is, therefore, much larger than the assumed distance of 3 cm. This calculation suggests that we cannot expect a significant fraction of the radon progeny to fully thermalize before traveling the distance between the radon source and the copper. However, we can expect their velocities to be reduced by collisions with air molecules in the vessel, and we can expect that an electric potential below 60kV might still stop a measurable fraction of radon progeny before they can plate-out on the copper. We used these calculations to guide our choice of electric potential for conducting the real experiment, described in the next sections.

5 Experimental Setup

As mentioned in previous sections, camping lantern mantles from before the 1990s are used as the primary source of ^{220}Rn within the setup. The decay of ^{220}Rn often yields alpha particles through alpha and beta emission, which is what is measured as the source of background. Figure 3 shows the camping lantern mantles in their assigned positions for the setup. The activity of each mantle is not known, therefore, we were careful to label each of the eight mantles and always place them in the same position on the bottom of the pressure cooker. This assured the same exposure conditions for each experiment.

Figure 3 – Camping lantern mantles in position for experiment within the pressure cooker

A pressure cooker is used as the exposure chamber. Pressure cookers are inexpensive and provide a sealed environment to prevent leakage of radon from the vessel. This helps isolate the mitigation technique as the cause of any reduction in radon plate-out. The pressure cooker was fitted with hardware for pressure control, electrical connectors to apply high voltage within the system, and other input and output ports as needed. The system was leak-checked after modifications were made, and its integrity verified. Figure 5 shows the pressure cooker with modifications.

Figure 5 – Pressure Cooker setup with modifications for input and output

The pressure cooker shown allows for electrical connectors to input high voltage into the system. High voltage connectors from Gas Electronic manufacturer with a rating of 100kVDC were purchased and used in the system to allow for proper insulation and connections for a source with the necessary voltage. This connector is shown in Figure 6 as it is within the setup.

Figure 6 – High voltage connector within the pressure cooker setup

A 3D-printed copper holder was designed since it is electrically insulating from both the grounded pressure cooker and the metal fabric used to place high voltage around the copper. The design of the holder went through many iterations to be sure of the best use of the space in the exposure chamber. The holder was designed to support up to six 4 inch by 4 inch copper pieces, so that it fits well into the pressure cooker and allows for the high voltage lead to be at an appropriate distance from the high voltage fabric connector to reduce arcing (dielectric breakdown of the air). With the six-inch diameter of the pressure cooker, a 4.5 wide copper holder allowed for this fit. The 3D-printed copper holder is shown in Figure 7.

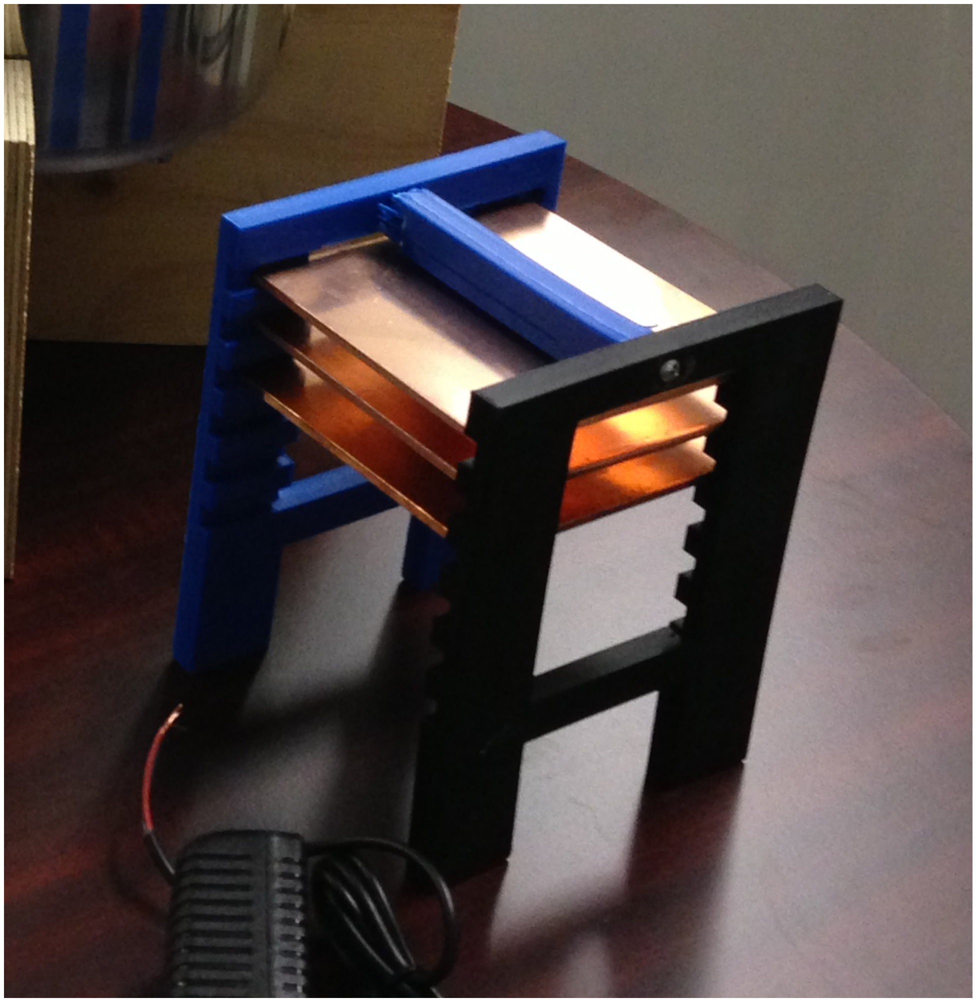

Figure 7 – 3D Printed Copper holder with copper plates used in experiment

Nickel-Copper (NiCu) fabric was purchased to use as a cap for the electrical connection inside the pressure cooker to surround the copper samples The cap can then be biased to the desired high voltage level with respect to the grounded pressure cooker. The NiCu fabric covers five of the six sides, leaving the bottom open. However, this still produces an electric field configured to repel positively charged radon daughters (or attract negatively charged daughter to the NiCu cap) as shown in Fig. 8. The field model shows the positively charged radon daughters would move towards the walls of the pressure cooker as desired, making the fabric use desirable since it fits tightly around the designed holder.

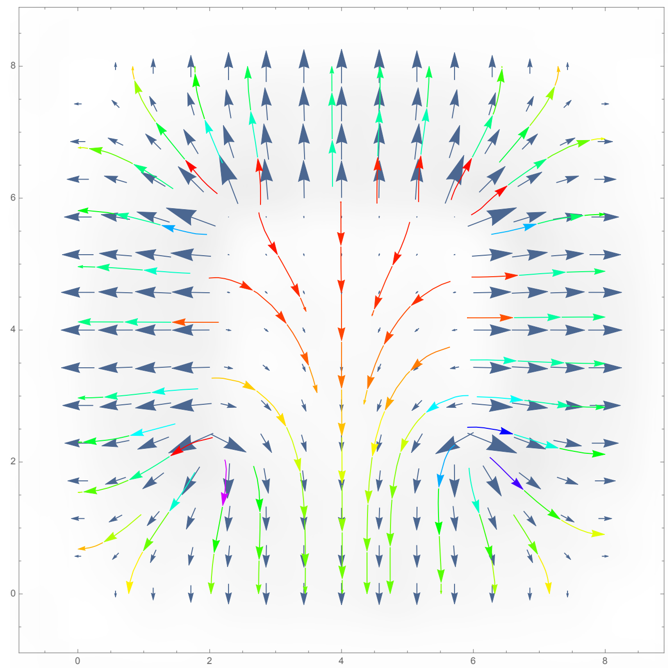

Figure 8 – Simplified model of electric field inside the pressure cooker

The NiCu fabric was sewn with common plastic fishing line to provide an insulated bond between panels of NiCu fabric in the cap, while guaranteeing that the panels themselves make good electrical contact with one another and represent a continuous electrical surface. The conductivity of the cap was verified by measuring the electrical resistance between the top and sides of the cap; all sides made good contact and we observed very low resistance across the cap. Figure 9 shows the copper fabric fit over the 3D-printed copper holder.

Figure 9 – NiCu fabric being placed over the 3D-printed copper holder

The copper holder is placed inside the pressure cooker with the NiCu fabric over it as shown. Alligator clips with insulation are then used to make good electrical contact between the cap and the high-voltage lead to the fabric. This setup is shown in Figure 10.

Figure 10 – Connection setup to transfer voltage from the high voltage lead to the fabric

A control experiment was performed prior to the electric field experiment to verify exposure levels of radon within the system. The NiCu cap was used in the control experiment in case it had some independent effect on preventing radon plate-out even with the field off. For each test, a five-day exposure was used to reach appropriate levels of measurable radon. The system was setup exactly as with the proposed electric filed on experiment to ensure the only variable was the application of the field.

Initial tests of the setup with a variable direct-current high-voltage poser supply (HVPS) suggested that arcing would occur between the corners of the cap and the pressure cooker walls at about 6-8 kV. This set the maximum voltage we could hope to use with the configuration Even without arcing, we detected some corona effects that resulted in production of ozone. Concerned with the health implications of producing ozone in a laboratory environment, the first operation of the electric shield was conducted in a fume hood in the SMU chemistry student laboratories. The HVPS was purchased from Information Unlimited due to its ability to produce up to 35 kV and its relatively inexpensive cost.. The initial setup is shown in Figure 11.

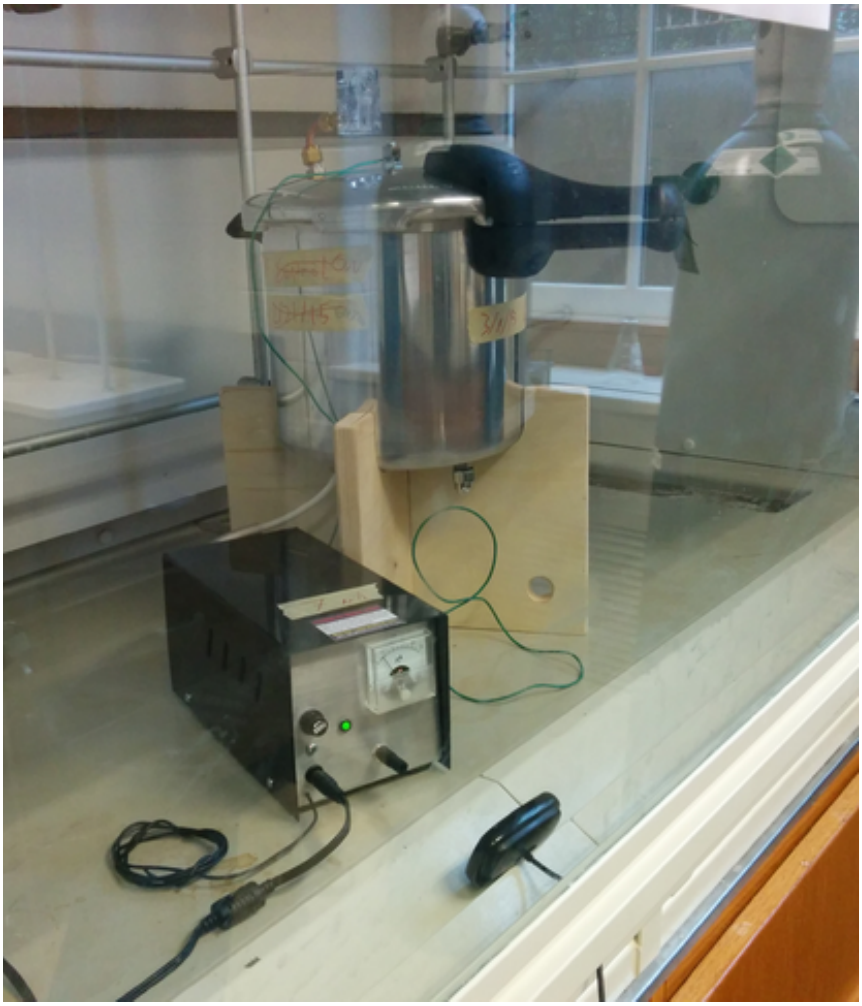

Figure 11 – High Voltage test setup in fume-hood with power supply shown

A confirmation experiment was done with the same voltage applied as first field-on, 6 kV. Different researchers rotated duties to ensure no results were caused by researcher-specific handling issues the equipment or the copper plates after the exposure.

6 XIA UltraLo 1800

Contamination levels were inferred be measuring alpha particles from the ^{220}Rn decay chain using the XIA Ultra-Lo 1800 instrument. This instrument is a specialized ionization counter comprising an active volume filled with argon, a lower grounded electrode that is a conductive tray holding the sample and an upper pair of positively charged electrodes, shown in Fig. 12. Of these two electrodes, the anode sits directly above the sample, while the guard electrode surrounds and encloses the anode. Both electrodes are connected to charge-integrating preamplifiers whose output signals are digitized and then processed by a digital pulse shape analyzer. The inherent background in the counting chamber of the XIA is 0.001 alphas/(cm^2 h).

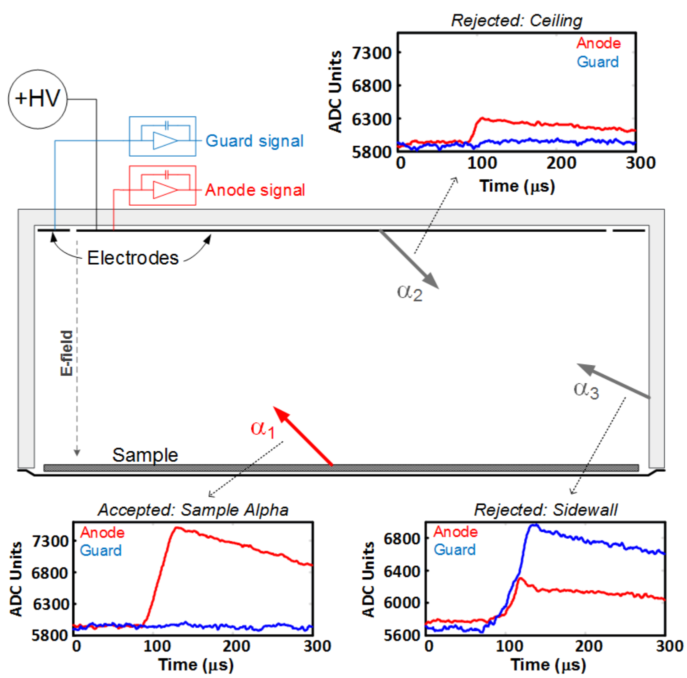

Figure 12 – XIA UltraLo 1800 Schematic

Figure 13 shows a typical view of the analysis and operations software used by the XIA, Counter Measure. This software is used to show the fingerprint of energies left by alpha particle emission of decaying isotopes, and allows us to see the amount of each isotope present within the copper samples. With the amount of alphas and the emissivity of the samples assessed out in real time, analysis can be done to verify expected levels of energy produced by the proper decay chain. These results are used to track levels of radon within the system, and can show any reduction of radon plate-out onto the copper surfaces.

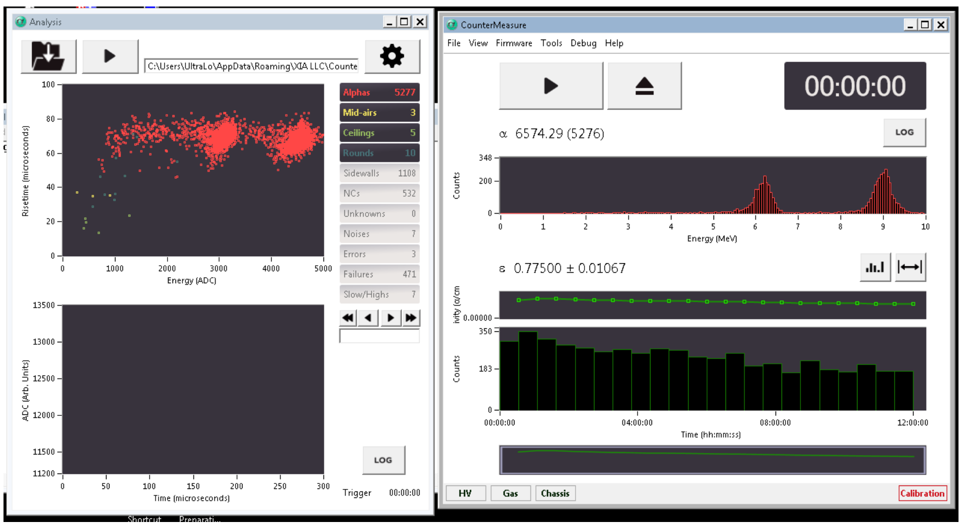

Figure 13 – CounterMeasure software from the XIA UltraLo 1800

7 Results

The results display an extremely promising reduction in background levels. We observe a reduction in 98.1% of radon levels plating-out on copper shown in Table 1.

Table 1 – Results from the control and experiment

Experiment	Emissivity 40 h after exposure (alphas/(cm^2*h))
E-Shield/Field Off	15.9 +/- 0.2
E-Shield/Field On	0.45 +/- 0.04

Even though the experiment only ran with one-tenth of the desired electric field, the results showed significant levels of reduction in the background in the experiment. The confirmation experiment showed an even better reduction, roughly by a factor of 2. While an improvement over the original field-on exposure trial, these results are statistically inconsistent despite attempting to hold conditions constant. This inconsistency is discussed in the next section. Figure 12 shows a plot of the emissivity as a function of time for the three experiments performed in this study. The electric field control and experiment are shown on the graph as labeled.

This set of experiments demonstrated a clear and quantifiable reduction in radon plate-out on copper using a specific, large electric potential.

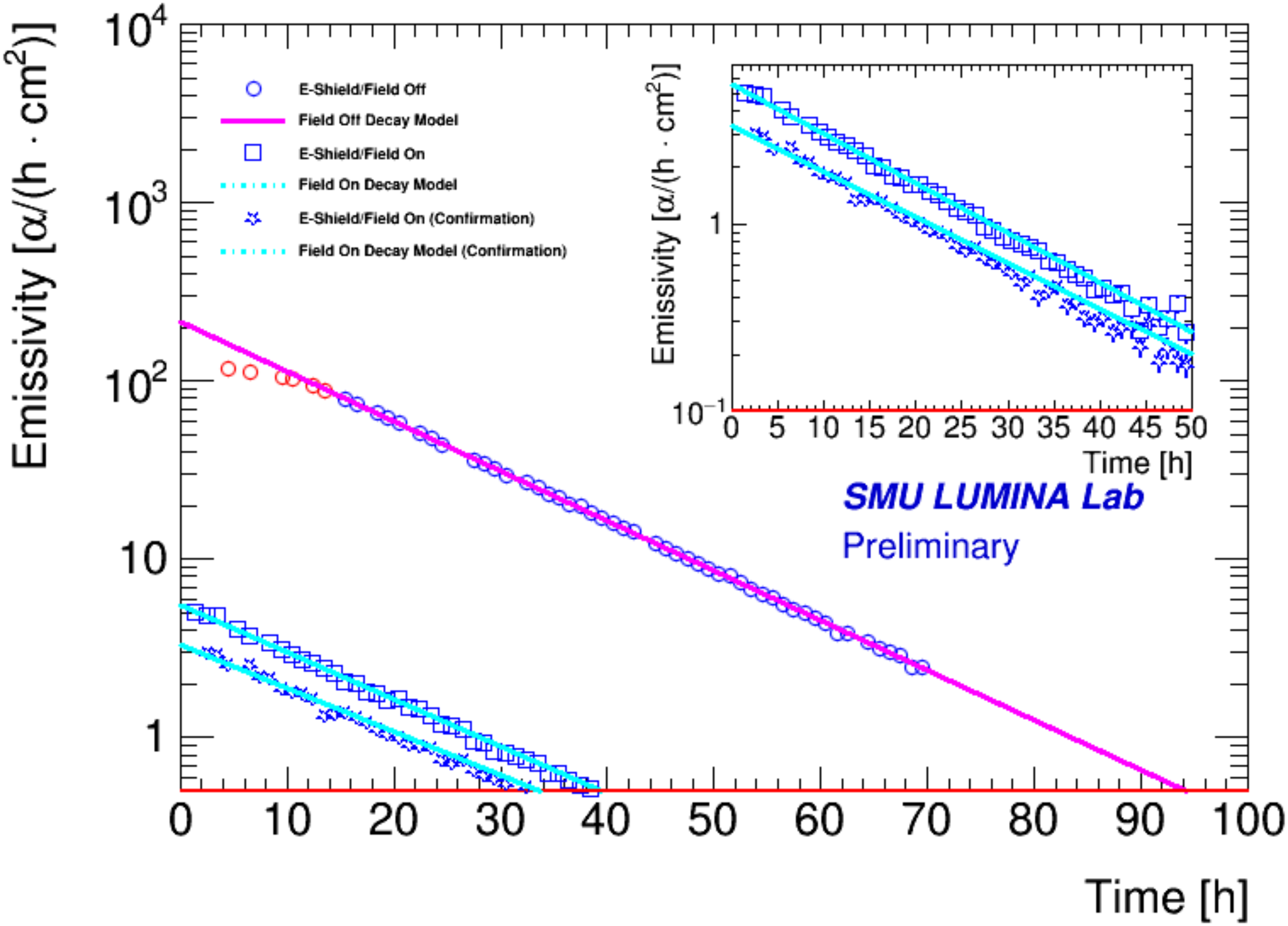

Figure 14 – Results for the control experiment, initial electric field experiment, and the confirmation experiment

8 Conclusions

The goal of this experiment was to quantify the effect that strong electric fields might have on reducing the plate-out of radon progeny on materials. We determined through some simple calculations that, in the worst-case scenario, a stopping potential of 60kV was required to completely prevent radon progeny from reaching the target materials. We designed an experiment to test this, isolating the effect of the electric field. Due to experimental constraints such as arcing, we were only able to operate at 6kV and not the maximum possible operation of 35kV that the power supply provides. Nonetheless, we observed a 98.1% reduction in radon plate-out with the electric field powered on in the setup.

As mentioned at the end of the last section, we ran a confirmation experiment to access the reliability of the electric field's ability to reduce plate-out of radon. We found a factor of about 2 improvement over the original experiment. The exact cause of this is unknown, and requires further study and investigation in a better-controlled environment. First, the two field-on experiments were conducted in slightly different external environments: one in a fume hood, the second in the normal laboratory environment. We observed that ozone degraded the rubber seals around the electrical and plumbing ports in the vessel in both experiments. For a future experimental design, we will replace the off-the-shelf rubber o-rings and seals with a product known to resist reactions with ozone degradation. For a further iteration of the experiment, we may look into alternate designs for the E-shield setup that will isolate the stored materials from ozone.

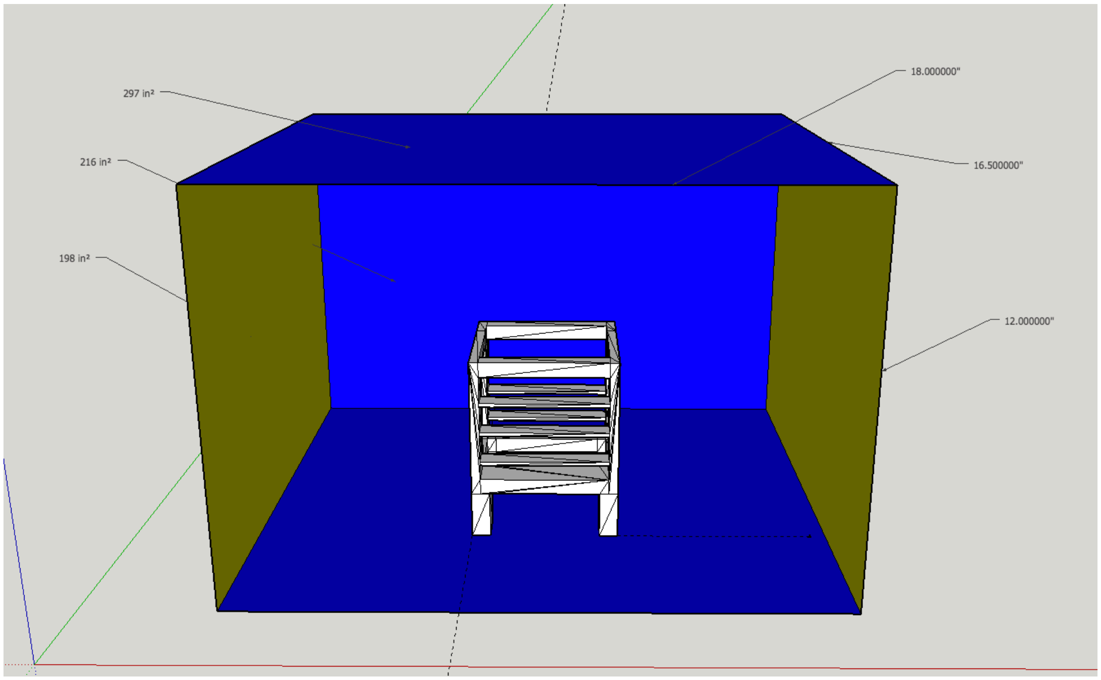

Figure 15 – Schematic for potential redesign of experimental setup

Figure 15 shows a potential redesign to replace the pressure cooker for the experimental vessel used in the E-shield experiment. The vessel is designed with the experiment in mind, and makes use of predetermined insulated surfaces, shown in blue, and conductive surfaces, shown in yellow. The electric field can be applied to the NiCu fabric as previously done, but the ground can be applied to the two conductive surfaces in hopes of attracting the radon daughters to these surfaces for counting purposes. These conductive surfaces can be removed from the experiment so that the radon measurements before and after the experiment can be determined for better understanding of the experiment. The dimensions are designed to be able to use the full 35kV from the HVPS without arcing. Tests were done to ensure that 6 inches is plenty of separation so that no arcing occurs in the experiment throughout the exposure period.

Experimental setup and calculations were hugely successful in bringing together a cohesive experiment that allows a test for the community of low background research to better understand storage and shielding techniques for reducing radon plate-out on copper surfaces. Future experiments will help us to further quantify exactly how exactly electric fields contribute to repelling radon and its daughter particles away from setups. Future projects for other undergraduate research students are already in the planning stages, starting with the projects outlined above.

Valuable experience was gained in analyzing and properly setting up high voltage electric fields. This information and conclusions on the ability for electric fields to repel radon progenies will allow further investigation into what has been decades of research in this field. The author is grateful for all those that have helped him in his course of study and those who will continue into the investigations as to how to improve low radioactive studies using electric fields to reduce background.

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