

Low Background Techniques 2004 Proceedings Surface Contamination From Radon Progeny

Michael Leung

Princeton University Physics Department, Princeton NJ 08544

Abstract. This paper examines methods the Borexino experiment uses to reduce surface contamination from radon progeny. The first section describes the deposition of airborne radon progeny. It includes measurements of the deposition rate in clean room conditions and describes steps that were taken to minimize the exposure of critical components in the experiment. Part two covers the adsorption of contaminants on surfaces through liquid contact. It summarizes measurements of the partitioning constant, cleaning effectiveness, and the time scales for the desorption of contaminants from a surface.

Keywords: Borexino, solar neutrino, low background, surface contamination, radon progeny, adsorption, deposition

PACS: 01.30.Cc

INTRODUCTION

The sensitivity of physics experiments looking for neutrino oscillations, dark matter, and neutrinoless double beta decay is limited by radioactive backgrounds. These backgrounds are often difficult to distinguish from real events and must be suppressed by many orders of magnitude for a successful measurement to be made. The source of radioactivity arises from either bulk impurities in the materials, external sources, or surface contamination. This paper focuses on surface contamination that results from gas and liquid adsorption of radon progeny. It also discusses steps that are taken to minimize these surface impurities. The studies are directed towards meeting high purity requirements in the Borexino experiment.

The Borexino solar neutrino experiment is designed to measure the ^7Be solar neutrino flux[1]. The detection method is neutrino-electron scattering in 100 tons of organic liquid scintillator. Taking into account the measured oscillation parameters, Borexino is expected to observe about 35 neutrino events per day in an energy window from 250-800 keV. Since it is desirable to have a large signal to noise ratio, the total rate of background events in this volume must be less than several per day from all sources. Achieving material and scintillator purity that meet this requirement is the main technical challenge facing the experiment. A diagram of the Borexino detector is shown in figure 1.

RADON

One of the most prevalent radioactive backgrounds is from radon gas which is naturally present in air. Typical indoor radon activities are $10\text{-}100\text{ Bq/m}^3$ though they can increase to several thousand Bq/m^3 in mines and caves where low background experiments are usually located to shield cosmic rays. The decay chain of radon is shown in figure 2. The half life of radon is 3.8 days but it is continually replenished by emanation from the ground where deposits of uranium reside. To reduce radon activity, buildings are typically ventilated with clean outside air.

Radon is particularly difficult to control due to its high mobility. In practical terms this means low background detectors must be designed to be leak tight to air. As an additional constraint, surface contact of detector materials with radon progeny must also be minimized.

Properties of radon and its progeny have been extensively studied to understand human radiation exposure from their inhalation, which is a cause of lung cancer. Many of the associated measurements and models are also of interest to low background experiments that must deal with surface contaminants. An excellent review of the literature can be found in *Radon And Its Decay Products In Indoor Air* by W. Nazaroff and A. Nero [2].

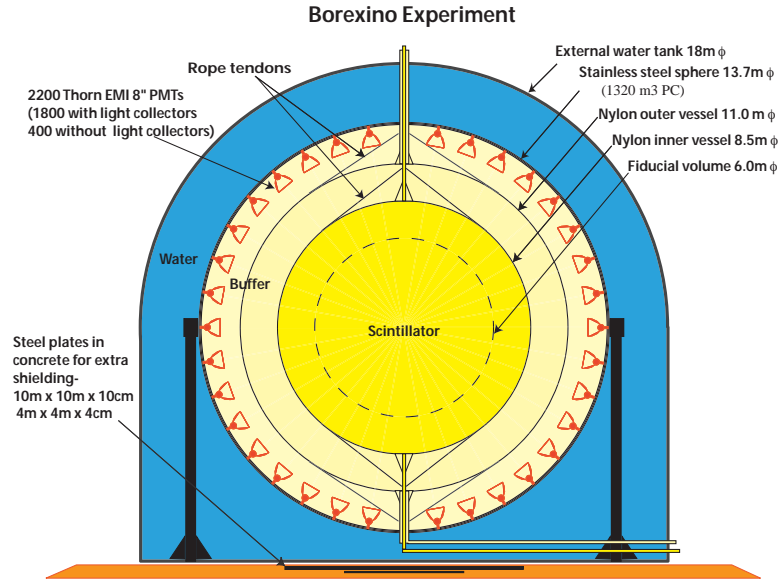


FIGURE 1. A diagram of the Borexino detector. The sensitive volume of the detector is a liquid scintillator solution composed of 1,2,4-trimethylbenzene (pseudocumene) with 1.5 g/l of 2,5-diphenyloxazole (PPO), a wavelength shifter. It is shielded by an inactive buffer of pseudocumene with dimethyl 1,2-benzenedicarboxylate (dimethylphthalate) which quenches scintillation. Events are observed with 2200 8" PMTs mounted on a stainless steel sphere. A high purity water shield outside the stainless steel sphere acts as a muon veto.

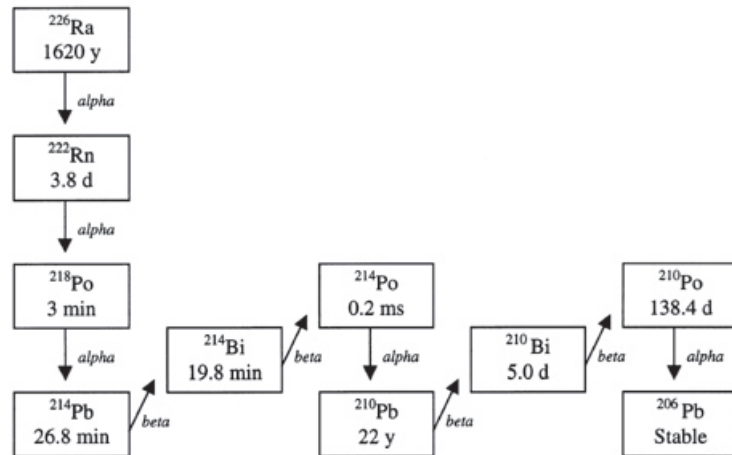


FIGURE 2. The radon decay chain. Radon is produced directly from the α decay of radium. When radon decays it produces a series of progeny with short half lives of less than an hour until ^{210}Pb is formed. ^{210}Pb with a half life of 22.3 years produces ^{210}Bi (β decay, $Q = 1.17$ MeV) and ^{210}Po (α decay, $Q = 5.3$ MeV) before reaching the stable isotope ^{206}Pb . It is the long half life of ^{210}Pb and the high energies of the two subsequent decays which makes radon exposure such a problem for Borexino.

DEPOSITION OF RADON PROGENY

Though radon is a noble gas and remains airborne, its progeny readily stick to surfaces. Over time these progeny build up ^{210}Pb which has 22 year half life. Since this half life is long compared to the lifetime of a typical experiment it is infeasible to wait for ^{210}Pb to decay away. The ^{210}Pb produces ^{210}Bi and ^{210}Po which β and α decay respectively, adding to the radioactive background. Thus the deposition of radon progeny on surfaces results in the presence of three radioactive isotopes long after the surface is isolated.

Radon progeny are formed in energetic decays allowing them to easily ionize and react with molecules the environment. The exact chemistry is difficult to predict as it is strongly dependent on the concentrations of various pollutants in the atmosphere.

The concentration of radon progeny in a room is relatively constant when measured sufficiently far from the walls. Near the walls, the concentration of the progeny decreases because the walls act like a sink. The surface concentration σ is the product of flux J towards the walls and the time of exposure t , for a stable element. The flux is found from the diffusion equation

$$J(x,t) = -D\nabla C(x,t) \quad (1)$$

where D is the diffusion constant and $C(x,t)$ is the concentration at a given position and time. However, if the concentration is constant in time and space (away from surfaces) it is convenient to instead represent the flux J as the product of the room concentration C and a deposition velocity v_d . The deposition velocity is the diffusion constant D divided by the boundary layer l near the surface

$$v_d = \frac{D}{l} \quad (2)$$

The boundary layer l is the region near a surface where the air is essentially static and convection is negligible. Thus the deposition velocity can be thought of as the rate at which particles diffuse towards a surface. The deposition of radon progeny can then be described by

$$\sigma(^{210}\text{Pb}) = \sum_{i=1}^5 C_i v_d t \quad (3)$$

where:

$\sigma(^{210}\text{Pb})$ = the surface density of ^{210}Pb atoms [atoms/m²];

C_i = the concentration of i^{th} radon daughter [atoms/m³];

i = index denoting specific progeny $\{1, 2, 3, 4, 5\} \rightarrow \{^{218}\text{Po}, ^{214}\text{Pb}, ^{214}\text{Bi}, ^{214}\text{Po}, ^{210}\text{Pb}\}$;

v_d = the deposition velocity [m/s];

t = the exposure time [s];

The ^{210}Pb does not appear immediately but is produced when the short lived precursors that stick to the surface decay. Since the time scale of low background experiments is on the order of years, the time for these quickly decaying isotopes to become ^{210}Pb can be safely neglected.

Studies have found that for progeny unattached to aerosols the deposition velocity $v_d = 4 \times 10^{-3}$ m/s in an average room[3]. This number can fluctuate by a factor of two in either direction depending on the local conditions. Usually the deposition velocity is measured for a particular situation since it depends strongly on variables which are not easily determined.

The deposition velocity is strongly influenced by the attachment of progeny to aerosols, which reduces it by roughly a factor of 10. Generally the fraction of progeny attached to aerosols increases with aerosol density though it is also dependent on the size distribution of the aerosols. A partial list of the factors influencing the deposition velocity is given in table 1.

The concentration of each radon progeny is determined by its half life, the concentration and half life of its parent, the ventilation rate of the room, the surface area of the room which acts a sink, filtration, and surface recoils which can free attached progeny. The Jacobi Model[4] (which is not discussed here) uses these parameters to describe the steady state progeny concentrations with a set of coupled differential equations.

Deposition Measurements

The nylon vessels in Borexino remain in direct contact with the liquid scintillator and are the nearest component to the fiducial volume making a high level of vessel cleanliness essential. Measurements were performed to test the build up of ^{210}Pb on a nylon surface in a clean room environment[5]. A small test clean room (2 m×2 m×3 m) was built to simulate the full scale class 100 clean room where the Borexino nylon vessels were fabricated. A clean room environment differs significantly from a typical room because the aerosol concentration is small enough that the attachment of radon progeny is negligible. The air flow due to the filters is also significantly higher than normal. Though the clean room filters do little to remove radon gas, they remove the radon progeny quite efficiently. It is worth

TABLE 1. Factors influencing the deposition velocity. A higher air velocity reduces the boundary layer near a surface, increasing v_d . Surface orientation is also a factor. Electric forces on charged progeny (a fraction of which are produced as positive ions when they decay) can substantially increase the deposition velocity.

Variable	Deposition Velocity
Air Velocity	Increased
Aerosol Attachment	Decreased
Charged Fraction	Increased*
Negative Ion Density	Decreased†
Surface Roughness	Increased

* if static charges are present on surfaces

† reduces charged fraction

noting that the test clean room was different from a standard clean room in that it was well sealed to retain radon during the exposure. This resulted in a small ventilation rate while maintaining a high filtration rate.

To measure the effective deposition velocity, nylon film samples (5 cm×5 cm) were placed in the test clean room. A 2MBq radon source was used to expose the room and nylon samples to a high radon concentration. The radon concentration in the test clean room was monitored over time using a RAD7 radon detector. The samples of nylon were then removed and the ^{210}Pb surface contamination was measured. The actual measurement was of the ^{210}Po α decays with a silicon detector. The ^{210}Pb rate was calculated by accounting for the build up time of ^{210}Po . Since only the radon concentration was measured and not that of the progeny, the ^{210}Pb surface contamination is expressed as

$$\sigma(^{210}\text{Pb}) = \left[\sum_{i=1}^5 C_i \right] v_d t = [kC_0] v_d t \quad (4)$$

where we take the sum over progeny to be proportional to the initial radon concentration C_0 . The constant k which is a small number dominated by the high filtration rate, is incorporated into an effective deposition velocity $v_0 = kv_d$. The ^{210}Pb surface contamination is then

$$\sigma(^{210}\text{Pb}) = C_0 v_0 t \quad (5)$$

where C_0 is the radon concentration and v_0 is an effective deposition velocity. The test clean room experiment provided the integrated radon exposure and ^{210}Pb surface contamination. It showed that in a clean room environment the effective deposition velocity is

$$v_0 = 3 \times 10^{-8} \text{ m/s} \quad (6)$$

This effective deposition velocity is roughly three orders of magnitude smaller than the average deposition velocity for an unattached radon progeny in a typical room. The most likely explanation is that the high filtration rate removes the radon progeny from the air before they can reach the nylon surface. If this is indeed the case then the effective deposition velocity should scale with the filtration constant λ_f which is the volume normalized filtration rate (number of volumes filtered per time).

Borexino Nylon Vessel Fabrication Precautions

The Borexino nylon vessels were fabricated in a customized class 100 clean room in Princeton, NJ. This clean room used a radon filter to provide low radon air for ventilation. The filter was a vacuum swing adsorption system which could supply up to 170m³/h of air with a radon activity of <0.3 Bq/m³[6]. This clean room was sealed to minimize the make up air required to keep an over pressure and to prevent the back diffusion of radon. Inside the clean room the radon activity was reduced from ~30 Bq/m³ to 1 Bq/m³ during fabrication. The clean room specifications are show in table 2.

TABLE 2. Specification for the class 100 clean room where the Borexino nylon vessels were fabricated. Modifications were made to reduce the deposition of radon progeny. The filtration constant $\lambda_f = 0.036/s$

Parameter	Value
Volume	770 m ³
Filters	84 HEPA
Flow Type	Laminar
Relative Humidity	50%
Temperature	290 K
Ventilation Rate	0.024 m ³ /s
Filtration Rate	28m ³ /s
Particulates $d > 0.5 \mu\text{m}$	< 3530/m ³
Radon Activity	1 Bq/m ³

During the vessel fabrication the nylon film was discharged with static rods to prevent charged progeny from being attracted to the vessel. It was also kept well covered when not in use. Only the edges where the joints were made saw significant air exposure. Since the nylon vessels will be filled with water for many weeks during commissioning, it is expected that much of the deposited contamination will be removed. Details can be found in reference [7].

ADSORPTION

Data from the Counting Test Facility (CTF), a small scale prototype of the Borexino detector initiated the groups interest adsorption from liquids. The CTF uses a nylon vessel with a one meter radius that is filled with the same liquid scintillator as Borexino. It is surrounded by a water shield and PMTs that observe the scintillator. The total CTF surface activity on the nylon vessel is several times the total internal activity of the scintillator. The surface activity has shown a slow decay over time roughly consistent with ²¹⁰Pb and that is uncorrelated with changed in the bulk activity.

Adsorption describes processes where molecules from a gas or liquid bind to a surface. Adsorption is an exothermic process that is broadly classified as either physical, involving Van der Waals binding; or chemical, where chemical bonds are formed. Physical adsorption is a reversible process that approaches an equilibrium where as chemical adsorption is an irreversible process often involving the dissociation of molecules.

A flat surface typically has a surface binding site density $n^s = 10^{-5}$ moles/m² or 6×10^{18} sites/m². This number reflects how many molecules can be attached in a monolayer in a given surface area. When a surface is in contact with a liquid all of the binding sites are occupied. The mole fractions of species on a surface N_i^s are generally different from the species' mole fractions in the liquid or gas.

The following describes physical adsorption at the interface of a binary solution and a solid surface. The analysis is simplified when the solution is dilute[8]. Let the solvent be component 1 and the solute be component 2. The moles of solute n_2^s adsorbed per area is related to the mole fraction of solute on the surface N_2^s and the adsorption sites n^s per area by

$$n_2^s = N_2^s n^s \quad (7)$$

The equilibrium constant for adsorption to a surface is

$$K = \frac{N_2^s a_1}{N_1^s a_2} = e^{-\Delta G^o / RT} \quad (8)$$

where a_i is the concentration of component i in solution and the N_i^s are the mole fractions on the surface. ΔG^o is the adsorption free energy. Since the solution is dilute a_1 is constant and

$$N_2^s = \frac{n_2^s}{n^s} = \theta = \frac{ba_2}{1 + ba_2} \quad (9)$$

where we define $b = K/a_1$ and use the fact that $N_1^s + N_2^s = 1$. θ is the fractional surface coverage of the solute. If ba_2 is much less than one then the mole fraction on the surface is simply proportional to the concentration in the liquid.

TABLE 3. ^{210}Pb removal from various cleaning methods on stainless steel. These results are representative of the relative efficiencies of each method. They are also indicative of the desorption rate for each process.

Soak Treatment	Temperature	Time	Percent Remaining
Pseudocumene	20° C	10 Hours	96.1±1.1%
Pseudocumene	20° C	1 Day	95.9±0.8%
Water	50° C	1 Day	63.7±0.9%
Burklin Detergent	50° C	1 Day	16.0±0.6%
Burklin Ultrasonic	20° C	1 Hour	11.7±0.5%

Partitioning Measurements

When pseudocumene is delivered for the experiment it contains relatively high concentrations of contaminants including ^{210}Pb . Before it can be purified the pseudocumene comes in contact with pipes, storage tanks, and column packing material. These stainless steel surfaces have an area of hundreds of square meters. This steel had the potential to adsorb a significant fraction of the contaminants. These surface contaminants could later be released into the pseudocumene after it had been purified.

A laboratory test was done to determine the equilibrium constant for the partitioning of ^{210}Pb between an electropolished stainless steel surface and pseudocumene. Pseudocumene was exposed to a high concentration of radon gas which was allowed to decay producing ^{210}Pb . The electro polished stainless steel sample (3 cm×3 cm) was then soaked in the spiked pseudocumene (~500 ml). The activity of the ^{210}Pb in the pseudocumene and on the stainless steel sample was then measured to find K . Two 2" photomultiplier tubes were used to measure the ^{210}Po content of the scintillator using a pulse shape discrimination technique from which the ^{210}Pb concentration was derived. The steel sample was counted with a planar germanium detector which observed the 46 keV ^{210}Pb gamma.

The results of the measurements are as follows. The measured ^{210}Pb rate of 36 kBq/m³ in the scintillator implied a lead concentration $a_2 = 6.1 \times 10^{-11}$ mol/m³. The surface density of ^{210}Pb with a rate of 1.2 Bq/cm² is $n_2^s = 2.0 \times 10^{-11}$ mol/m². Thus the fraction surface coverage $N_2^s = 1.3 \times 10^{-6}$ and $N_1^s \approx 1$. The concentration of PC is $a_1 = 7.3 \times 10^3$ mol/m³, and stays essentially constant since the solution is dilute. Thus we find

$$K = 1.6 \times 10^8 \quad (10)$$

and $b = K/a_1 = 2.1 \times 10^4$ m³/mol.

Wash Off Measurements

The reaction rates at which a liquid and adsorbed phase approach equilibrium are not necessarily the same for adsorption and desorption. While contaminants are adsorbed quite rapidly, desorption is a much slower process. This has implication for various cleaning methods. A set of measurements was done to determine the time scale for desorption of ^{210}Pb on steel.

The results of the desorption measurements are shown in figure 3. The graph shows that the desorption appears to be a function of log time. This is unfavorable for cleaning processes that involve flushing or soaking pipes and tanks with water because removal of surface contaminants is slow. Measurements of the desorption rate of ^{210}Pb from nylon into water also show a logarithmic dependence on time, though the rate is faster. It is suspected that the desorption rate is generally logarithmic for radon progeny bound to flat surfaces. The rate is a function of the substrate, adsorbed species, contact fluid, agitation, and temperature.

Several measurements have been done to test the effects of aggressively cleaning and not cleaning steel samples with a detergent. Data are shown in table 3. With no detergent cleaning of lines and tanks, desorption is a problem for recontamination in Borexino. These measurements show that the desorption rate of ^{210}Pb from steel into pseudocumene is slower than desorption into water.

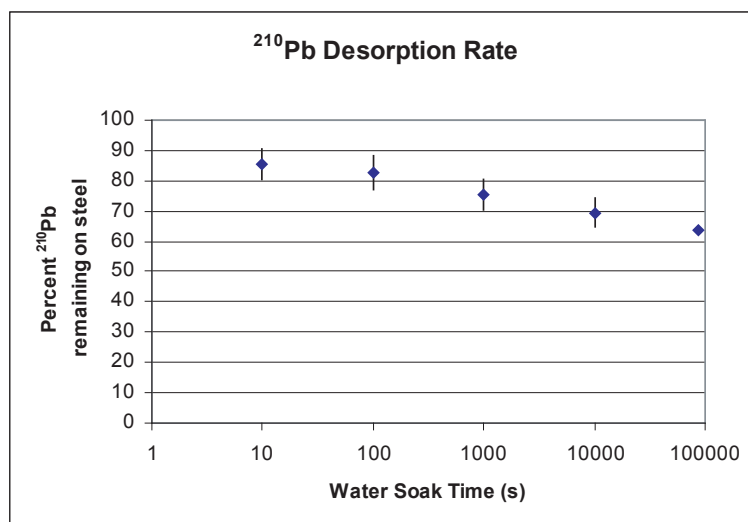


FIGURE 3. The desorption of ^{210}Pb from stainless steel into water as a function of time. The steel ($3\text{ cm}\times 3\text{ cm}$) was soaked in one liter of $10\text{ M}\Omega$ de-ionized water at 50° C . The first four data points were made with the same sample that was repeatedly counted and soaked. A separate sample was measured with higher statistics after a one day soak. The 10 second data point exhibited higher ^{210}Pb removal due to agitation during the water soak.

SUMMARY

Radon progeny are a source of contamination that low background experiments must contend with. ^{210}Pb in particular which is produced from ^{222}Rn has a long half life and is followed by two high energy decays before a stable isotope is reached. While bulk contamination of materials from radon and its progeny must be dealt with, adsorption of these impurities on surfaces can also be significant. When considering surfaces contamination in designing a low background experiment it would be useful to know the following:

- 1) The partitioning constants for radioactive elements in a liquid between the different surfaces they contact.
- 2) The desorption rates to determine potential recontamination and to understand the effectiveness of cleaning.
- 3) The radon concentration, effective deposition velocity and time of exposure during fabrication. Better would be the concentration of each progeny concentration, deposition velocity, and exposure time.

This information can be attained through small scale laboratory measurements if not previously available.

ACKNOWLEDGMENTS

I would like to thank Frank Calaprice, Jay Benziger, Cristian Galbiati, Tom Shutt, and Kevin McCarty for their help with the measurements. This work was made possible by National Science Foundation Award Number 002961-001.

REFERENCES

1. The Borexino Collaboration, G. Alimonti et al., *Astropart. Phys.*, **16**, 205-234, 2002.
2. W. Nazaroff and A. Nero, *Radon And Its Decay Products In Indoor Air*, John Wiley & Sons Inc., New York, 1998.
3. G. Schiller, *Ph.D. Thesis*, University of California Berkeley, Princeton NJ, 1984.
4. W. Jacobi, *Activity and potential alpha energy of ^{222}Rn and ^{220}Rn daughters in different air atmospheres*, *Health Physics*, **22**, 441, 1972.
5. T. Shutt, *Radon Plateout Program*, Internal Borexino Memo, Princeton NJ, 2000.
6. A. Pocar, *Ph.D. Thesis*, Princeton University, Princeton NJ, 2003.
7. A. Pocar, *Low background techniques for the Borexino nylon vessels*, Low Radioactivity Techniques 2004 proceedings, 2005.
8. A. Adamson and A. Gast, *Physical Chemistry of Surfaces*, John Wiley & Sons Inc., New York, 1997.