Emanation and Diffusion of Radon through Gaskets for SuperCDMS SNOLAB

Richard Schnee,[1,a)] Brandon DeVries,[1] and Michael Bowles[1,2]

[1)] *South Dakota School of Mines & Technology, Rapid City, SD, 57701-3901, USA*
[2)] *now at The Johns Hopkins University Applied Physics Laboratory, Laurel, MD, 20723-6099, USA*

[a)] *Corresponding author: richard.schnee@sdsmt.edu*

Abstract. The SuperCDMS SNOLAB experiment, currently under construction, will attempt to directly detect dark matter particles. Shielding surrounding the experiment's detectors will reduce interactions of particles from radioactivity and cosmic rays. A gas purge will remove radon from gaps in the shielding to reduce backgrounds further. Gaskets used to seal this purge volume must allow sufficiently low radon diffusion through them while emanating little radon into the purge volume. Radon diffusion, solubility, and permeability were inferred by measuring the time-dependent radon concentration in a volume separated by gaskets made of ethylene propylene diene terpolyme (EPDM), Zip-A-Way, or silicone. Although the silicone tested has better radon properties, EPDM also is sufficient and is easier to use, and so EPDM will be used for the SuperCDMS radon barrier, with ZIP-A-Way used to reduce diffusion further and patch leaks.

INTRODUCTION

The SuperCDMS SNOLAB experiment will shield high-purity silicon and germanium detectors within shielding depicted in Fig. 1 to reduce backgrounds in the search for WIMP dark matter [1]. Air gaps within the shielding would provide dominant backgrounds due to gammas from radon daughters ^{214}Pb and ^{214}Bi if the space were not purged with low-radon gas [2], due to the ambient cavern radon concentration $\lambda C_{\mathrm{SNO}} \approx 130\,\mathrm{Bq/m^3}$ [3], where λ is the radon decay rate and C_{SNO} is the cavern radon concentration in units of atoms/m^3. A requirement on the concentration within the shield $\lambda C \leq 1\,\mathrm{Bq/m^3} \equiv \lambda C_{\mathrm{req}}$ ensures that such radon produces less than 2% of the expected total background. To achieve this concentration, a purge flow $F \approx (1-10)\,\mathrm{L/min}$ of liquid nitrogen (LN) boil-off with an expected a radon

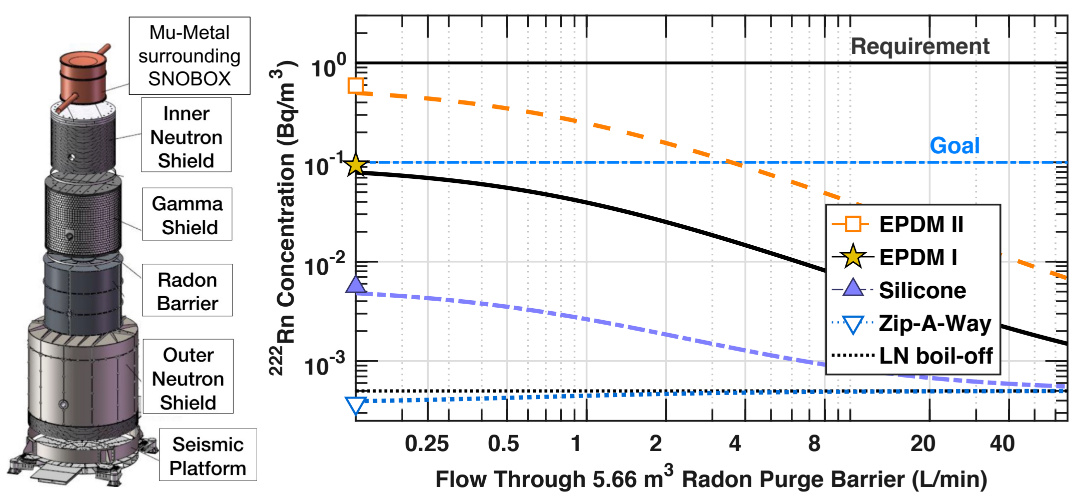

FIGURE 1. Left: Exploded view of the originally planned SuperCDMS SNOLAB shielding, with the radon barrier surrounding the inner shielding including 5.66 m^3 of air gaps outside of the mu-metal shield surrounding the SNOBOX. The additional 3.2 m^3 of air between the mu-metal shield and the SNOBOX is expected to be stagnant and so may be ignored in this paper. **Right:** Radon concentration within the radon barrier as a function of flow rate F resulting only from radon emanation of the gasket materials, assuming contribution $\lambda C_{\mathrm{LN}} = 0.5\,\mathrm{mBq/m^3}$ from the liquid nitrogen boil-off purge gas (dotted). The radon concentration requirement $\lambda C_{\mathrm{req}} = 1\,\mathrm{Bq/m^3}$ (horizontal solid) and goal $C_{\mathrm{goal}} \equiv 0.1\,C_{\mathrm{req}}$ (horizontal dashed) are shown. The zero-flow equilibrium concentration is shown (symbols indicated in the legend) on the y-axis for each of the gaskets measured. The radon emanation per length used here to determine the steady-state concentration corresponds to the 95% C.L. upper limit (2σ) for each gasket. Emanation results from both silicone and Zip-A-Way (triangles) are consistent with zero.

TABLE I. Summary of radon emanation and diffusion measurements for gaskets with 68% C.L. uncertainties or 95% C.L. upper limits. Radon emanation E for the material area emanated is translated into an expected emanation rate per unit length E_L using the gasket geometry or a 1-cm thickness for Zip-A-Way. Radon permeability P is listed for each material measured; only measurements of Zip-A-Way provided constraints on the radon diffusion coefficient D and solubility S. Inferred diffusion source term $\mathscr{D}$ for the assumed SuperCDMS SNOLAB geometry dominates over the emanation source term $\mathscr{E}$ for all materials measured.

Material	Area (cm^2)	E (mBq)	E_L (mBq/m)	$\mathscr{E}$ (Bq)	P (m^2/s)	D (m^2/s)	S	$\mathscr{D}$ (Bq)
EPDM I	252	2.9 ± 0.6	4.6 ± 0.9	0.24 ± 0.05	$5.8 \pm 0.1 \times 10^{-9}$	— no constraint —		6.8 ± 0.1
EPDM II	251	50 ± 4	52 ± 5	2.8 ± 0.3	— — — no diffusion measurement — — —			
Silicone	120	< 0.08	<0.29	<0.015	$2.0 \pm 1.2 \times 10^{-9}$	— no constraint —		2.3 ± 1.3
Zip-A-Way	54,000	< 0.09	<0.033	<0.0017	$2.5 \pm 0.1 \times 10^{-10}$	$2.2^{+1.0}_{-0.6} \times 10^{-10}$	$1.1^{+1.6}_{-0.7}$	0.29 ± 0.01

concentration $\lambda C_{LN} \lesssim 0.5\,\text{mBq/m}^3$ [4] will flush the gas within a hermetically sealed radon purge barrier surrounding the inner shield volume.

The air space between the radon barrier and the mu-metal shield has volume $V_S = 5.66\,\text{m}^3$. To allow easy construction and removal of the radon barrier, it is designed with 4 seams around the circumference and 4 seams separated by 90° running vertically, requiring a length $L = 53.0\,\text{m}$ of gaskets. Since most gasket materials have poor radon properties some care must be taken to ensure that the gasket material neither produces too much radon nor allows too much radon to diffuse from the cavern environment.

The radon concentration C inside the shielding decreases due to the purge and radon decay and increases due to the sources of radon emanation $\mathscr{E}$ and diffusion $\mathscr{D}$:

$$\frac{\partial C}{\partial t} = \frac{F}{V_S} C_{LN} - \frac{F}{V_S} C - \lambda C + \frac{\mathscr{E} + \mathscr{D}}{V_S}.$$

The steady-state radon concentration in the shielding (found by setting $\partial C/\partial t = 0$)

$$C = (FC_{LN} + \mathscr{E} + \mathscr{D})\,/\,(F + \lambda V_S)\,. \tag{1}$$

This concentration approaches the purge gas concentration C_{LN} for high flows. For practical reasons, it is desired that the flow not need to exceed 10 L/min. At such flow, the requirement that $\lambda C < 1\,\text{Bq/m}^3$ implies that $\mathscr{E} + \mathscr{D} < 85$ atoms/sec. At a (preferred) flow of 1 L/min, the requirement becomes $\mathscr{E} + \mathscr{D} < 13$ atoms/sec. Note that an emanation (or diffusion) source that produces N atoms/sec of radon would, if allowed to come to equilibrium so the decay rate equals the emanation (or diffusion) rate, result in an equilibrium number of atoms that produces a decay rate of N Bq. For simplicity (and as is often conventional), we will quote emanation and diffusion rates by their equilibrium decay rates, in units of Bq.

RADON EMANATION OF POTENTIAL GASKET MATERIALS

The South Dakota Mines radon emanation system [2, 5] was used to measure several potential gasket materials. Lemer Pax [6] provided two samples of self-adhering ethylene propylene diene terpolyme (EPDM) cellular foam gaskets from different production batches. EPDM began as the leading candidate to seal the shielding since it is self-adhering and thus easy to apply and remove, necessary during construction. They also sent a sample of silicone rubber. The other material tested was Zip-A-Way [7], a commercially available removable weather stripping manufactured by Red Devil. This material is designed to be applied as a beaded caulk in thin strips. We purchased several bottles for the radon diffusion and emanation measurements described here. The sample was prepared for radon emanation by emptying one full 300 mL bottle onto two sides of an aluminum foil of area $A_{foil} = 272\,\text{cm}^2$. The foil had previously been measured to emanate $< 70\,\mu$Bq.

Table I reports the radon emanation results of the potential gaskets. We see that EPDM gaskets may vary in their radon emanation depending on the production batch. The silicone and Zip-A-Way gaskets were consistent with zero radon emanation, with upper limits on their emanation lower than any other gasket materials published. The resulting emanation per length can be used to compute the level of radon added to the shielding from the gasket emanation. A gasket of length L with ^{222}Rn emanation per unit length E_L may contribute as much as $\mathscr{E} = E_L \times L$ to an enclosed

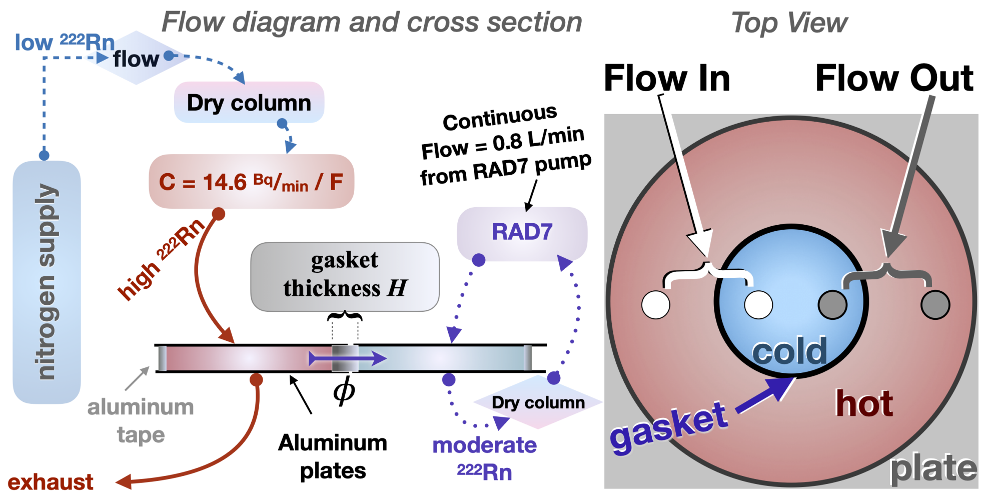

FIGURE 2. **Left:** Flow path diagram of the experimental apparatus to determine the radon diffusion properties (D,S,P) of potential gaskets. A gasket of thickness H separates two volumes sandwiched between two aluminum plates. The boil-off LN gas supply is regulated with a flow meter and fed to a high-activity radon source providing a near-constant output concentration on the 'hot side' (red), where the radon may be measured and exhausted. The radon diffuses through the gasket to the 'cold-side' (blue) of the apparatus, where the RAD7 is continuously circulating the diffused radon. A drying desiccant is placed inline to ensure a low relative humidity. **Right:** Top view of the apparatus (gray plate) with flow ports ('in' and 'out') on opposite ends of the hot (red) and cold (blue) sides to encourage thorough gas mixing to achieve a homogeneous radon concentration on each side.

volume. Although the actual amount is likely smaller by $\sim 2\times$, we conservatively assume the maximum possible emanation for analysis in this paper. The right panel of Fig. 1 shows the steady-state radon concentration due to emanation alone for SuperCDMS as a function of the purge flow.

RADON DIFFUSION THROUGH POTENTIAL GASKET MATERIALS

Radon diffusion was measured through the same potential gasket materials. The measurements used a custom experimental set-up sketched in Fig. 2, similar to *e.g.* those outlined in Refs. [8, 9, 10, 11]. The circular gasket under test divides two volumes formed within a pair of 12"-by-12" aluminum plates about 0.5 cm apart. The inner volume is prepared by initial purging with low-radon air and called the "cold" side. The outer volume, called the "hot" side, is sealed to prevent leaks to the lab and is supplied with air of high radon concentration C_{H} from a Pylon brand radon source [12] with activity 117 kBq. To obtain a known and approximately constant radon concentration, the source is first purged and then set with a constant flow $F \sim 2.5$ L/min, resulting in $\lambda C_{\mathrm{H}} \sim 6\,\mathrm{kBq/m^3}$. The radon from the hot side diffuses through the gasket under test to the inner, cold-side volume. The cold-side gas is dried with an inline desiccant to ensure low relative humidity and is continuously circulated with a commercial Durridge RAD7 radon detector [13], which measures the radon concentration as a function of time. For some runs, the hot-side concentration is confirmed by measuring with a RAD7. The total volume of the cold side and its tubing is kept small relative to the RAD7 internal volume to minimize systematic errors. For some measurements (not included here), a gasket did not prevent leaks of radon from the hot side to the cold side; such leaks were obvious from the fast equilibration of the cold-side concentration with that of the hot side [2].

The concentration $C(x,t)$ of radon in the (one-dimensional) gasket/membrane is modeled in one dimension as

$$\frac{\partial C}{\partial t} = D\frac{\partial^2 C}{\partial x^2} - \lambda C,$$

where D is the gasket diffusion coefficient. The concentration at the gasket-air interface is higher inside the gasket by a factor S known as the solubility. The resulting time-dependent radon concentration on the cold side of the gasket

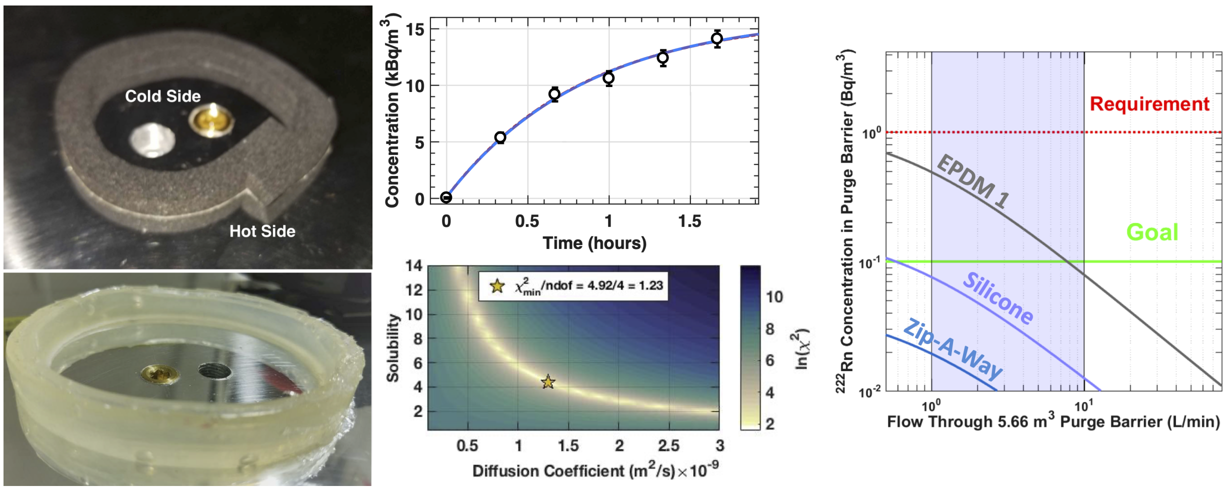

FIGURE 3. Left: EPDM (top) and Zip-A-Way (bottom) gaskets on aluminum plate before their diffusion measurements. The Zip-A-Way gasket has some air bubbles, which presumably increased radon diffusion moderately. **Top Middle:** Cold-side radon concentration versus time from an EPDM diffusion measurement (◯) with 99.7% C.L. statistical uncertainties. The best-fit theoretical expectation from Eqn. 2 (curve) has best-fit permeability coefficient $P_{\mathrm{EPDM}} = 5.8 \pm 0.1 \times 10^{-9}$ m^2/s. **Bottom Middle:** χ^2 (grayscale) as a function of EPDM solubility S and diffusion coefficient D showing the degeneracy between the two parameters for this dataset. **Right:** Expected steady-state radon concentration within the radon purge barrier versus flow rate F for the planned 15-mm-thick EPDM gasket (top curve), compared to expectations for silicone (middle) or Zip-A-Way (bottom) gaskets. All gaskets meet requirements (horizontal dotted) for all reasonable flow rates (shaded). The EPDM nearly meets the goal (horizontal solid) for the planned flow rate (vertical solid). The diffusion dominates the expected radon in the radon purge barrier.

depends on the gasket thickness H and exposed area A, and the cold-side volume V [9, 14]:

$$C(t) = C_{\mathrm{H}} \frac{ADS}{H\lambda V} \left[1 - e^{-\lambda t} + 2 \sum_{n=1}^{\infty} (-1)^n \times \frac{1}{1 + n^2 \pi^2 \cdot \frac{D}{\lambda H^2}} \left(1 - e^{-\beta_n t} \right) \right], \tag{2}$$

where

$$\beta_n \equiv \lambda + D \left(\frac{n\pi}{H} \right)^2 .$$

If $D/\lambda H^2 \gtrsim 1$, the steady-state concentration [2]

$$C(t \to \infty) \approx C_{\mathrm{H}} \frac{ADS}{H\lambda V}. \tag{3}$$

The steady-state concentration gives a good measurement of the permeability $P \equiv DS$ but cannot constrain the diffusion constant nor solubility independently. The diffusion coefficient D may be determined separately from the time dependence of the increase of the radon concentration, so long as the grow-in is sufficiently slow.

For the diffusion measurement, the second EPDM sample was cut into a strip of thickness $H_{\mathrm{EPDM}} = 2.7$ mm, as shown in Fig. 3, to avoid complications testing the original 15-mm thickness. A Zip-A-Way gasket, also shown in Fig. 3, was prepared by spraying between two KF flanges in a concentric configuration. The time-dependent radon concentration for the EPDM measurement is shown in the top-middle panel of Fig. 3. The grow-in was too quick to place satisfactory constraints on the diffusion coefficient D, but the permeability P is well constrained, as shown in the bottom-middle panel of Fig. 3. Two measurements of the silicone sample were made, but both had shortcomings potentially leading to high humidity [2]. The resulting systematic uncertainty on the permeability is about a factor of two and prevents constraints on the diffusion constant and solubility. Two measurements of Zip-A-Way were made [2], with the results of the second measurement suggesting a problem at early times that would result in underestimated permeability and diffusion. If early measurements are ignored, the results agree well with the first measurement, so those values are reported in Table I.

CONCLUSIONS

The radon source due to diffusion $\mathscr{D}$ within the SuperCDMS radon barrier can be found from Eqn. 3 using the planned SuperCDMS radon barrier gasket geometry, estimating the exposed gasket surface area $A = Lw$ with $L = 53\,\text{m}$ and the exposed width $w \approx 0.5\,\text{cm}$. For example, the EPDM gasket with radon permeability $P = 5.8 \times 10^{-9}\,\text{m}^2/\text{sec}$, with a thickness $H = 1.5\,\text{cm}$ gives a (zero-flow) equilibrium concentration

$$\frac{\mathscr{D}}{V_\text{S}} \approx \lambda C_\text{SNO} \frac{LwP}{H\lambda V_\text{S}} = 130\,\text{Bq/m}^3 \frac{53\,\text{m} \cdot 0.5\,\text{cm} \cdot (5.8 \times 10^{-9}\,\text{m}^2/\text{sec})}{1.5\,\text{cm} \cdot (2.11 \times 10^{-6}/\text{sec}) \times 5.66\,\text{m}^3} \approx 1.2\,\text{Bq/m}^3.$$

The right panel of Fig. 3 shows the expected steady-state radon concentration within the SuperCDMS SNOLAB radon barrier as a function of the purge flow for each of the three potential gaskets, based on Eqn. 1. For each gasket, Table I shows that the source of radon emanation $\mathscr{E}$ is much less than the radon diffusing $\mathscr{D}$ through a 15-mm gasket, due to the radon concentration $\lambda C_\text{SNO} = 130\,\text{Bq/m}^3$ of the SNOLAB cavern. All three gaskets meet the experiment's requirements even for modest flows.

During construction and commissioning, the SuperCDMS shielding will be opened and closed multiple times. The planned gasket will need to be easy to work with: application ought to be quick with little thought required, and removal should be easy as well. Both silicone and Zip-A-Way have better diffusion properties than the EPDM gasket, but the silicone lacks adhesive, and the Zip-A-Way dries too slowly except when applied in thin beads. We plan to use a self-adhering 1.5-cm-thick EPDM gasket to seal the radon purge barrier, with the Zip-A-Way caulking used to patch leaks and further reduce the radon permeation through the EPDM. After commissioning, it may make sense to replace the EPDM gaskets with one of the materials with better radon properties, to allow the radon concentration goal to be achieved with reduced (or even zero) purge flow.

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