

Measurements of Radon Diffusion and Emanation from Titanium

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Low Radioactivity Techniques Workshop 2026

September 23, 2026

**undergraduate*

Impact of Diffusion on Rn Emanation

- Two sources of Rn Emanation

- Diffusion of Rn out of material

- Follows Arrhenius relation.
 - Often negligible at cryogenic temperatures (~200 K).

- 86-keV Rn recoil from Ra α -decay

- Range is $\lesssim 0.1 \mu\text{m}$, so emanates only for surface contamination.
 - Often dominates, especially for cryogenic temperatures.

- Even at room temperature, can measure split between the two sources.

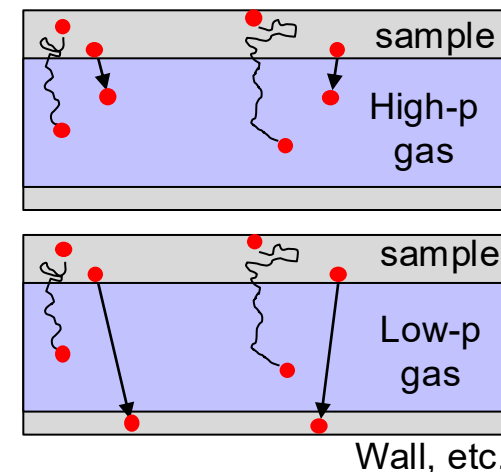
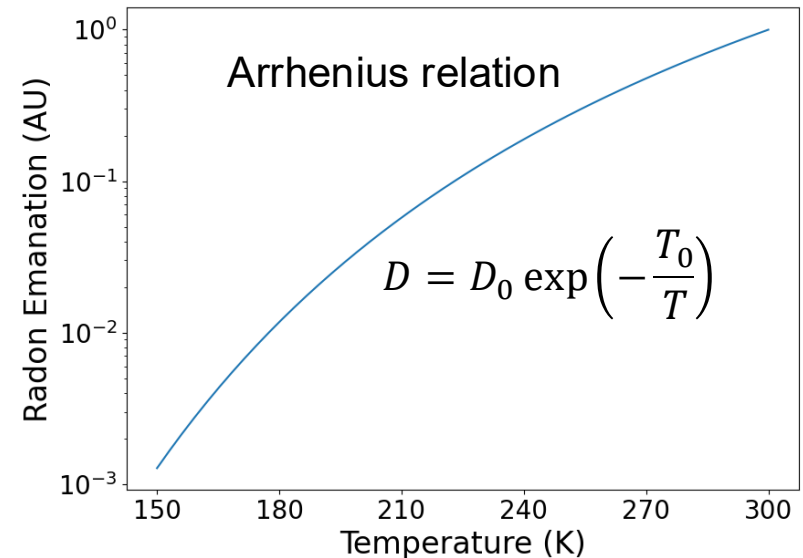
- Emanation into atmospheric-pressure gas collects both.

- Recoil range in gas $\sim 100 \mu\text{m}$, so $\sim$ all stop in gas (for smooth surface).

- Emanation into 100-mTorr gas collects $\sim$ only emanation from diffusion.

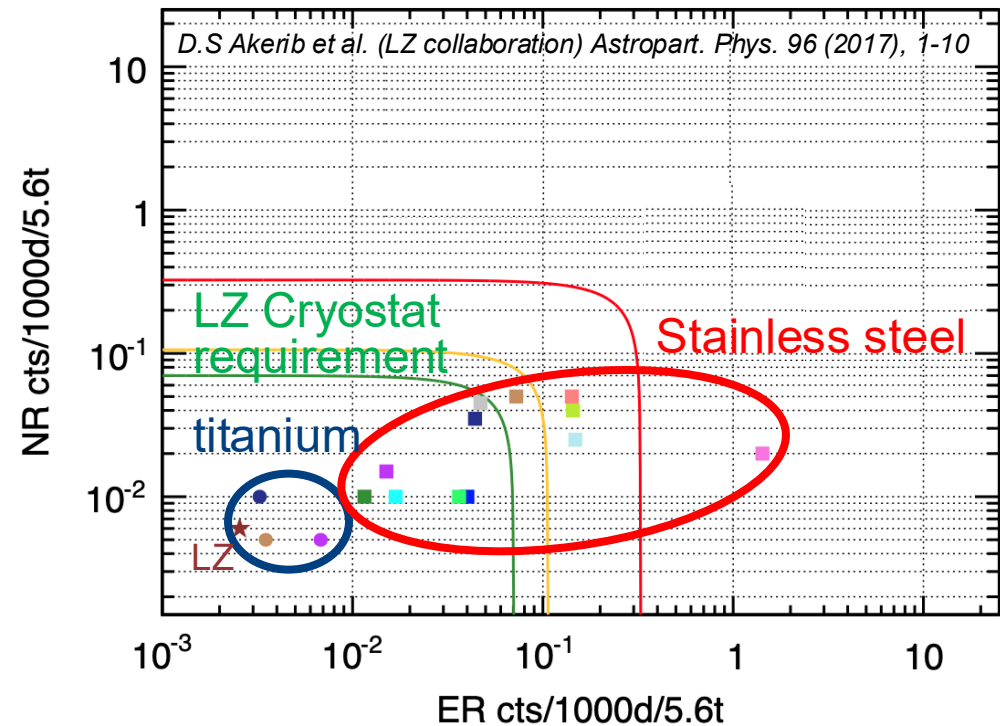
- Recoil range in gas $\sim 7 \text{ m}$, so $\sim$ all recoil-induced emanating atoms implant into another surface.

– Rn from recoil not collected.



Titanium for Low-Background Cryostats

- Excellent properties:
 - High strength-to-weight ratio
 - Lower-Z so less attenuation of gammas for better vetoing
 - Low cosmogenic activation
- Choice for LZ.
 - Lower backgrounds expected than for stainless steel (assumed 0.15 mBq Rn).
 - As-built measurements of LZ Ti Inner Cryostat Vessel (ICV) indicated ~100x higher Rn.
 - Largest source of Rn emanation in LZ, ~1 mBq/m² on inside.
 - Despite acid etching of surface and low ²²⁶Ra concentration in ICV (~4 mBq/m²).
 - Infer ~ 50% of radon emanates into inside or outside.



What went wrong?

How do we avoid such a problem in the future?
(Titanium favored for XLZD)

Possible causes of Rn Emanation from Ti

- Three possible causes:

1) Ra ~ uniform in Ti, but Rn diffusion length in titanium is much larger than typical for metals (microns)?

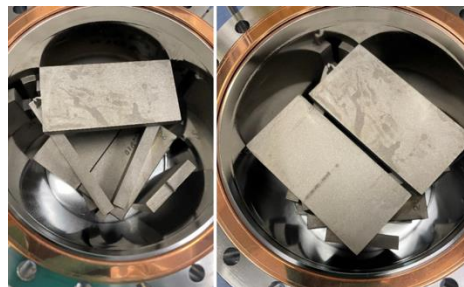
- Need diffusion length $\gtrsim 2$ mm even when cold.

2) Radium concentrated near surfaces but surface etching ~ineffective?

3) Contamination added during assembly (e.g. welds)?

- Impractical to rule out (3).
- Will discuss (1) and (2) in detail.
- E. Perry et. al. *JINST* **21** P09016 [arXiv:2607.22870](https://arxiv.org/abs/2607.22870) reported ~2x lower Rn emanation from cold Ti than from warm:

Temperature [K]	^{214}Po [mBq/kg]
293	41.14 ± 2.16
223	20.20 ± 2.17
188	19.72 ± 1.53



Lorenzo Ascenzo's talk yesterday says it is not diffusion in the Ti.

- Strongly suggests diffusion is responsible for ~half of Rn emanation at room temperature, shuts off when cold.

Measurement of Rn Diffusion through Titanium

- Was not aware of any previous measurements for titanium

- Use similar setup to those for previous measurements.

M. Wojcik, NIM B61, 8–11(1991).

M. Wojcik, W. Wlazo, G. Zuzel, & G. Heusser, NIM A449, 158 – 171 (2000).

M. Durcik & F. Havlik, J. Radioanal. Nucl. Chem. 209, 307–313 (1996).

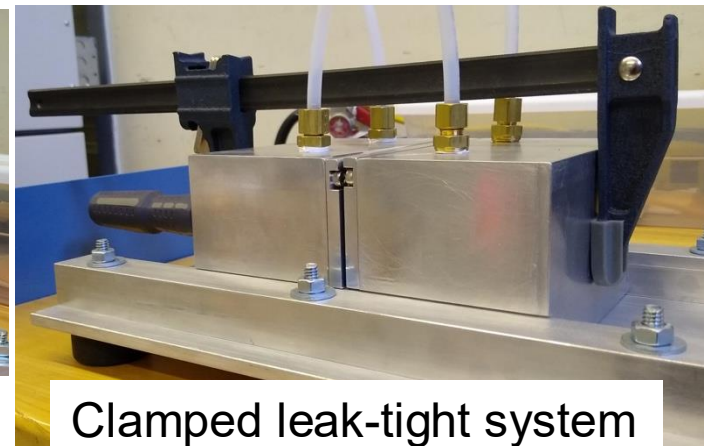
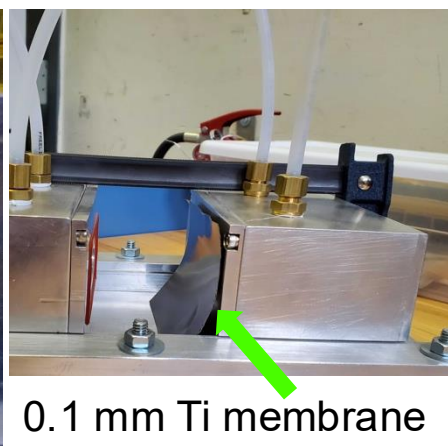
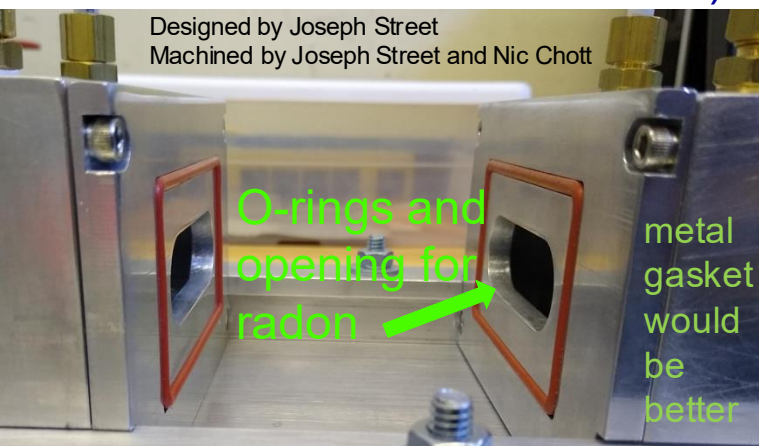
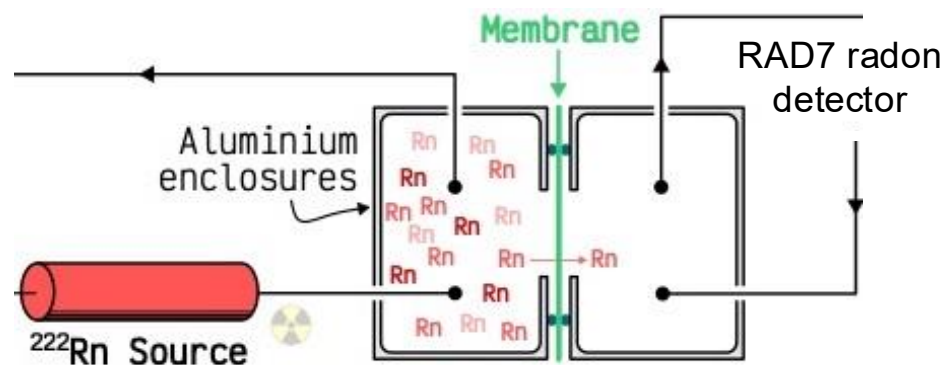
M. Jiranek & M. Kotrbat, Radiat. Prot. Dosim. 148, 274 (2012).

R.W. Schnee, B. DeVries, M. Bowles, *AIP Conf. Proc.* 2908, 100009 (2023).

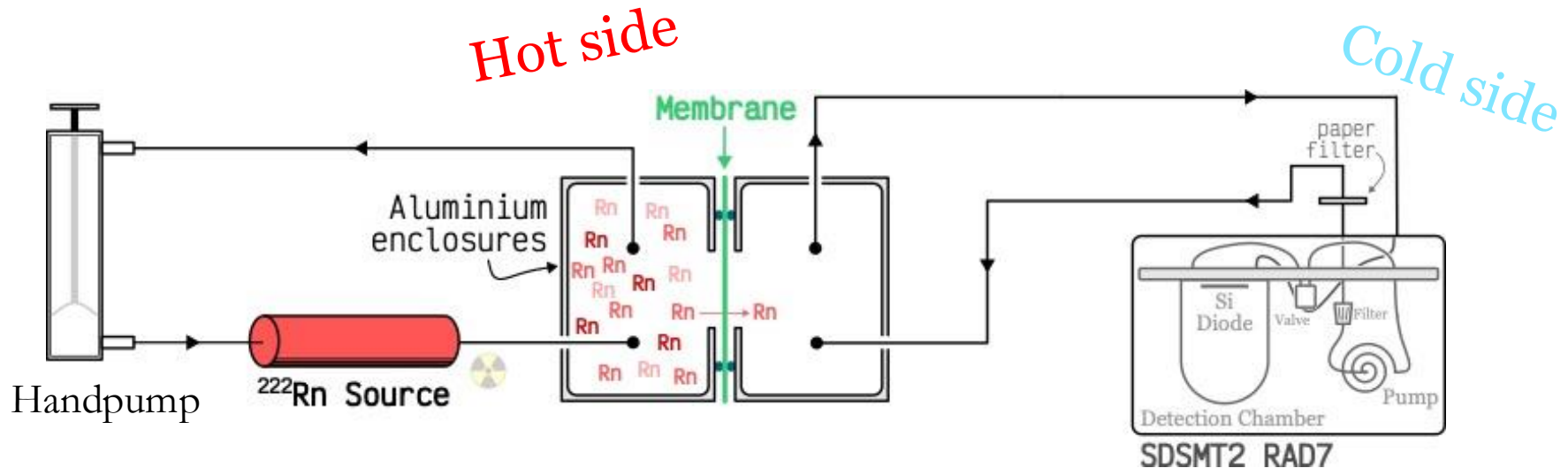
L. Wu et al. [arXiv:2410.06054](https://arxiv.org/abs/2410.06054) (2024).

- Undergraduate project.

- Home-made set-up.
- Put Rn on one side, measure diffusion to other, infer D and solubility S (= concentration in Ti / concentration in air).

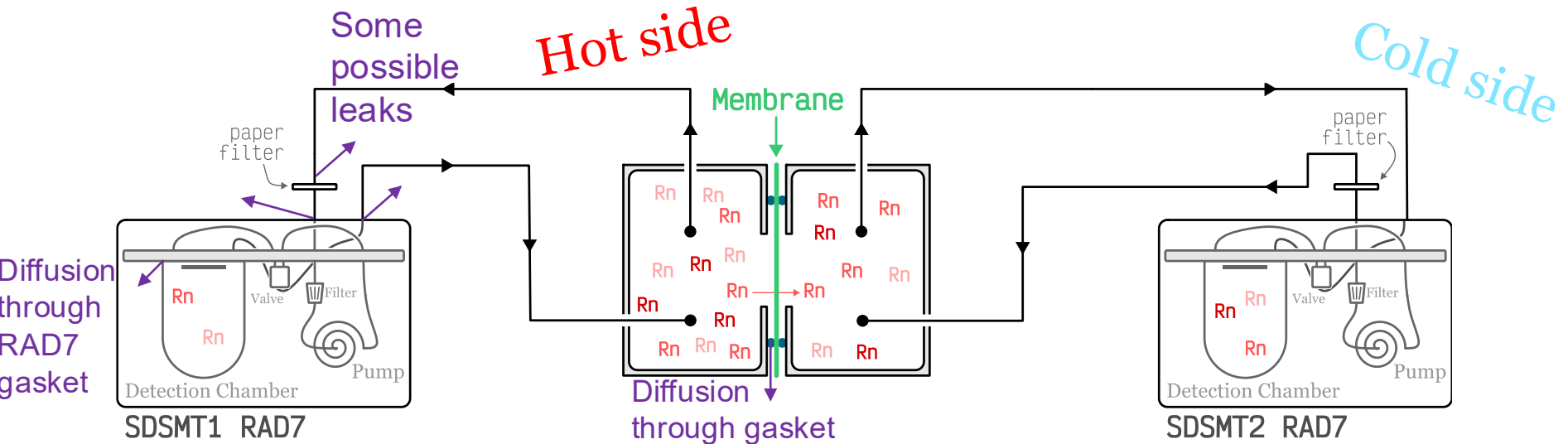


Measurement of Rn Diffusion through Titanium



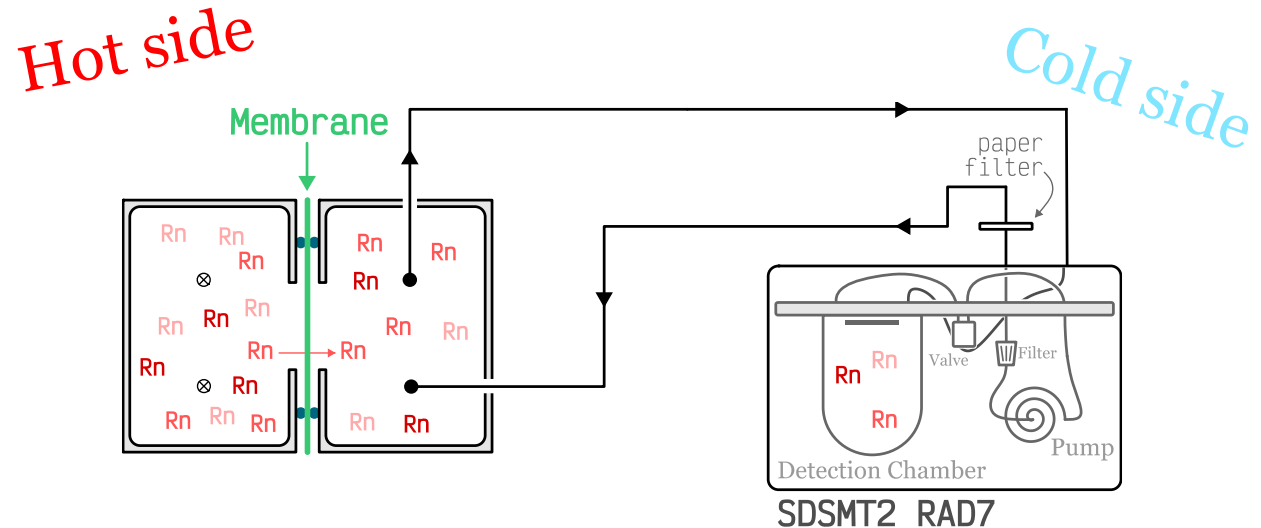
- Step 1: Add Rn to one (“hot”) side of chamber.
 - Connect Pylon radon source and hand pump to the hot-side system and mix radon throughout the hot-side volume.
- Step 2: Measure Rn on hot side of chamber.
 - Disconnect Rn source and pump. Connect RAD7. Measure Rn escape rate through RAD7 gasket + hot-side gasket + leaks.
- Step 3: Measure Rn vs. time on “cold” side of chamber.

Measurement of Rn Diffusion through Titanium



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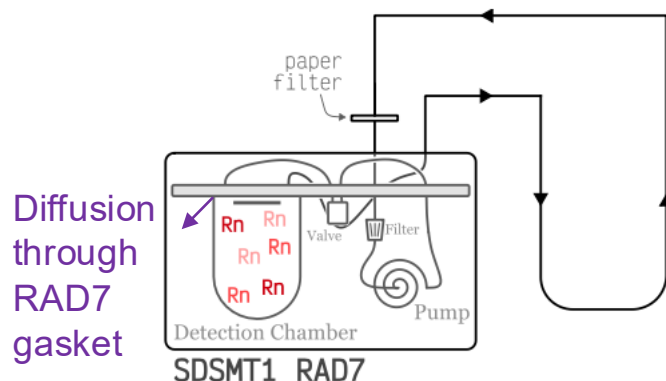
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*Make additional measurements to determine rates of **Rn** escape.*

RAD7 “leaks” & Response



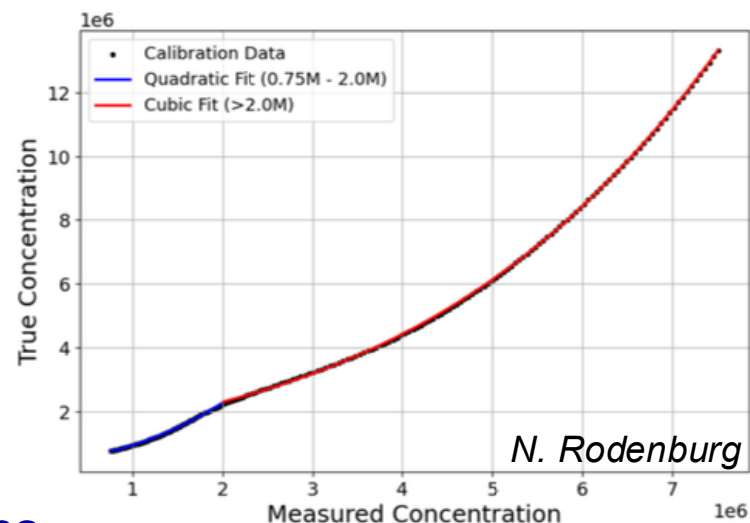
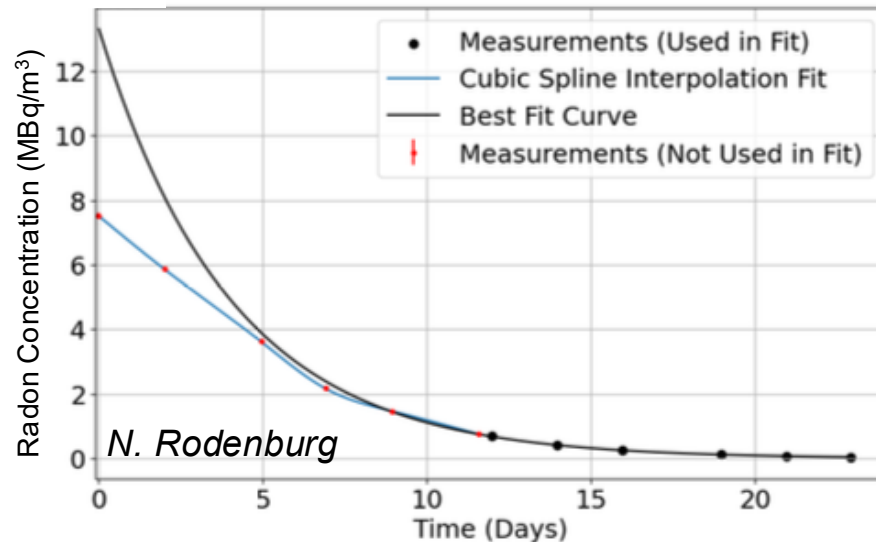
- Challenges:

- RAD7 response not accurate above 0.75 MBq/m^3 .
- RAD7 gasket allows radon diffusion from detection chamber.

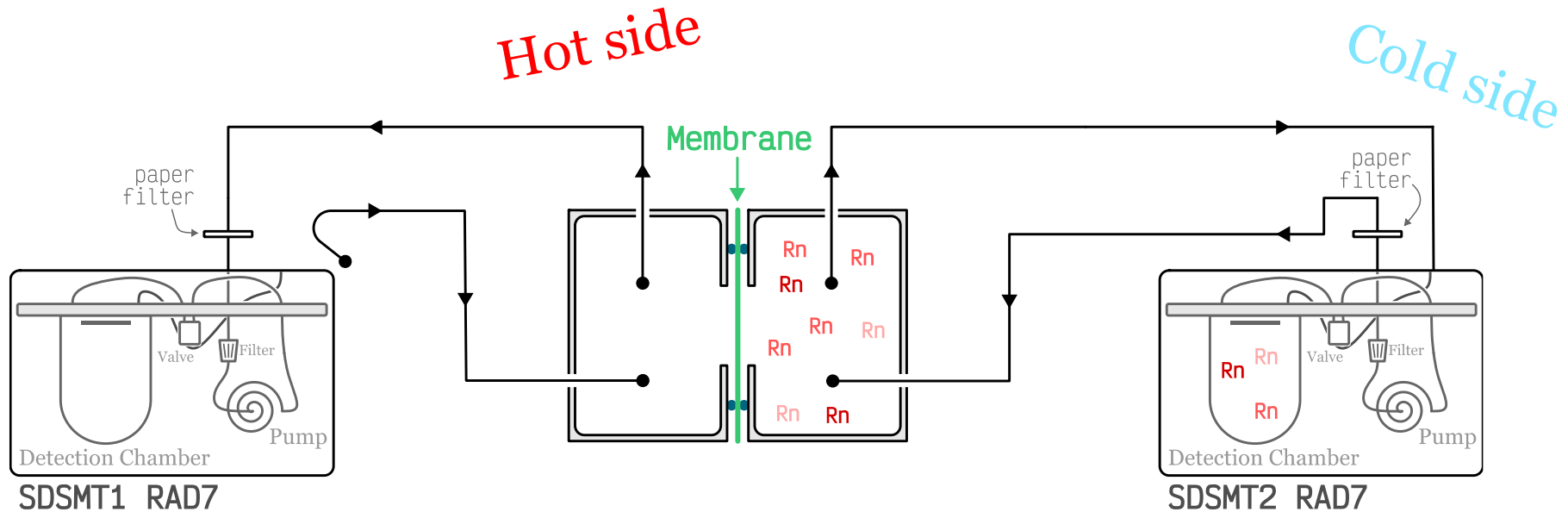
- Solution:

- Measure rate of radon leaving chamber using data $< 0.75 \text{ MBq/m}^3$.
- Extrapolate true inferred concentration to earlier times.
- Map True vs. Measured concentrations.

S.S. Poudel et al., arXiv:2606.00473 [quant-ph]

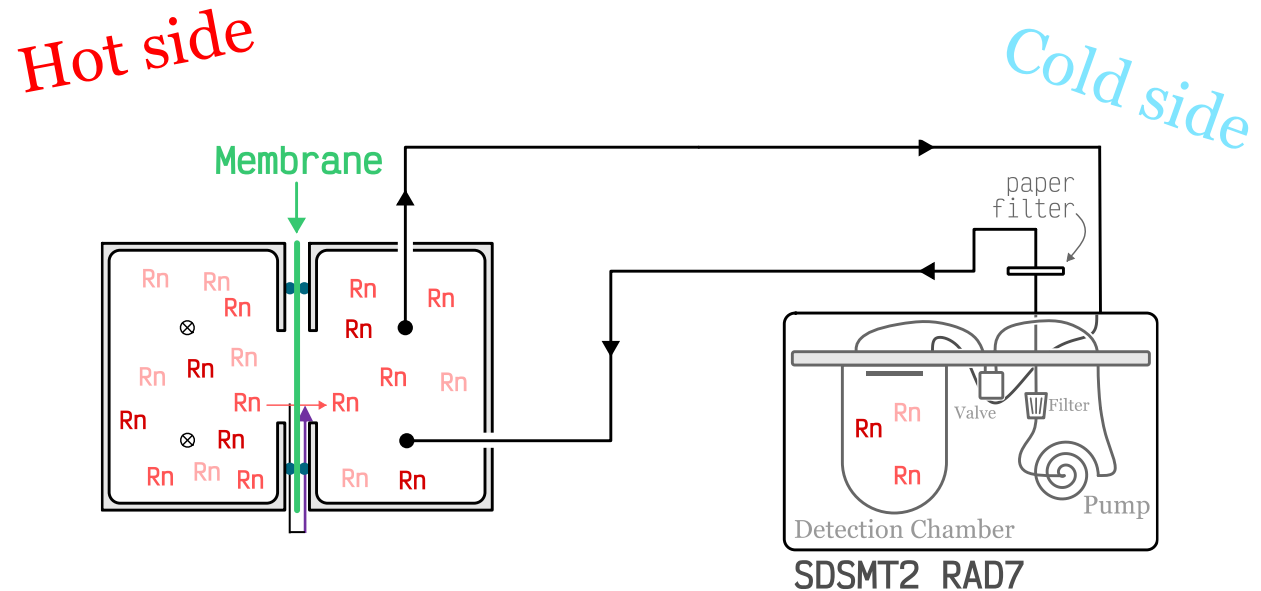


Other “Leak” Measurements and Concerns



- Also measure escape from cold side with no Rn on hot side.
 - Results consistent with escape from hot side.
- Possible shortcoming is radon from hot side exiting through gasket and entering cold-side through gasket.
 - Hope this is negligible, but results will provide upper limit on diffusion if such path is not negligible.

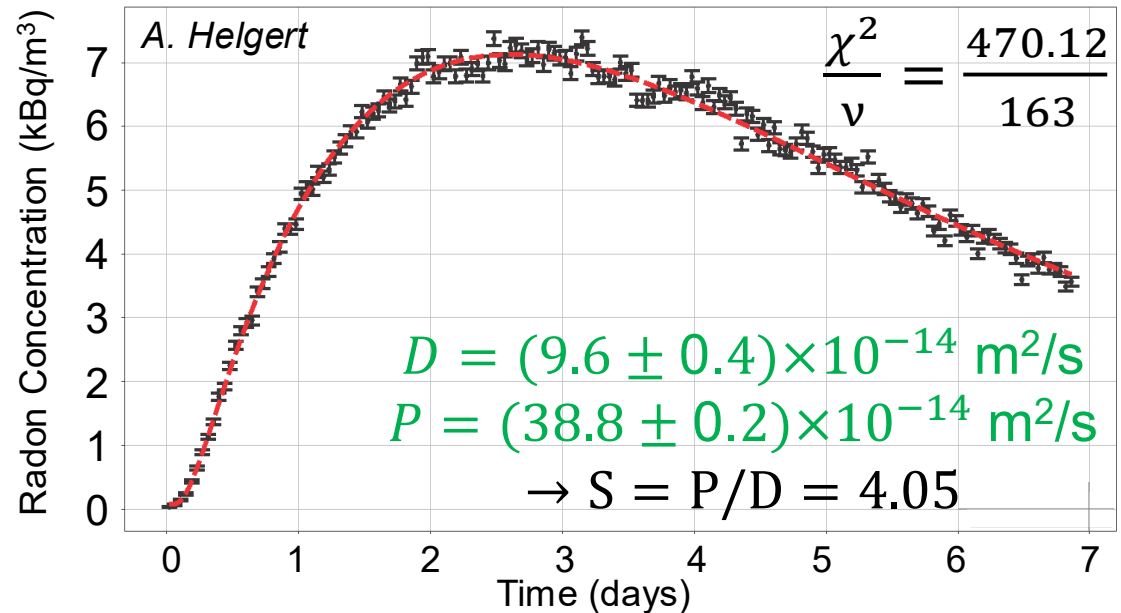
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Measurement of Rn Diffusion through Titanium

- Measure radon on cold side over time for 0.1 mm-thick Ti.
- Find best-fit parameters to numerical solution.

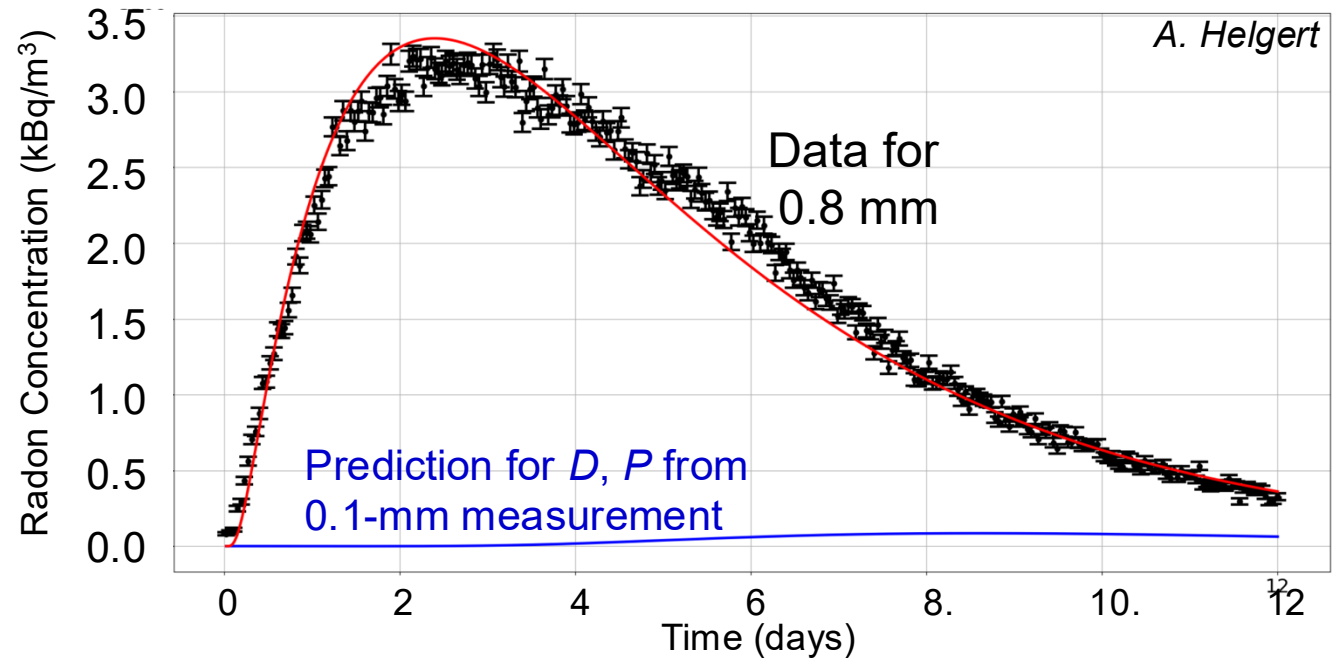


- Free parameters
Diffusion D and
Permeability P (= SD for Solubility S).
- Scatter from point to point is larger than uncertainties.
 - Uncertainties systematically underestimated – scale uncertainties on D , P .
- Although chi-square indicates poor fit, fit appears quite good by eye.
- Results imply diffusion length $l_D = \sqrt{D\tau_{\text{Rn}}} = 0.2 \text{ mm}$.
 - Most Ra in LZ cryostat Ti must be close to surface.

For confirmation of D , measure for different thickness (otherwise, this value of D is just an upper limit).

Upper limit on ~~Measurement of~~ Rn Diffusion through Titanium

- Performed measurement with 8x thickness (0.8 mm).



- Results not consistent with D & P from 0.1 mm.
- Must be dominated by leaks (= something other than diffusion through Ti) at 0.8 mm.
 - Likely dominated by leaks at 0.1 mm.
 - So previous measurement is an upper limit on diffusion of radon in Ti:
 - $D < 10^{-13} \text{ m}^2/\text{s}$, $P < 3.9 \times 10^{-13} \text{ m}^2/\text{s}$, Diffusion length $< 0.2 \text{ mm}$

Consistent with but less interesting/useful than Lorenzo's result.

Possible causes of Rn Emanation from Ti

- Three possible causes:

~~1) Ra uniform in Ti, but Rn diffusion length in titanium is much larger than typical for metals (microns)?~~

- ~~• Need length $\gtrsim 2$ mm even when cold.~~

2) Radium concentrated near surfaces but surface etching ~ineffective?

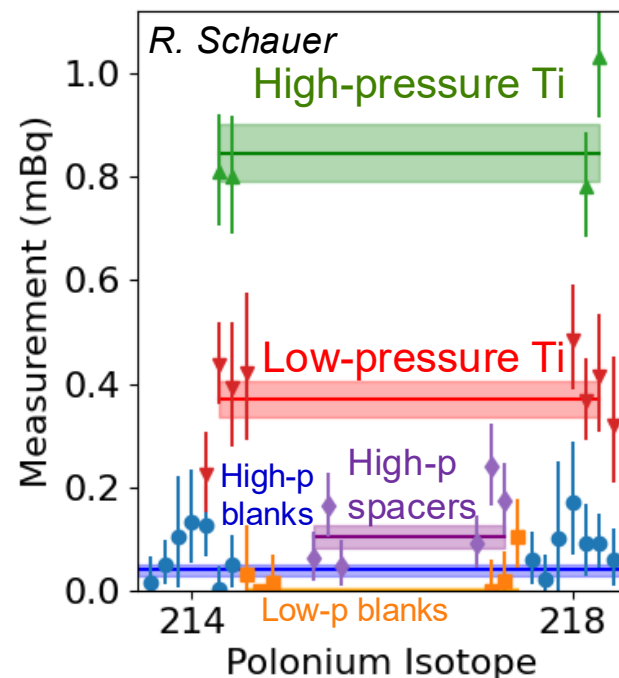
3) Contamination added during assembly (e.g. welds)?

- Impractical to rule out (3).
- Will discuss (1) and (2) in detail.
- XENON1T [E. Aprile *et al.* Eur. Phys. J. C 81, 337 (2021), arXiv:2009.13981] measured high ratio ^{222}Rn emanation to ^{226}Ra content (60%) in Ti, but
 - Electropolishing 30 μm reduced emanation by $>30\times$ to $\cong$ zero.
 - ^{226}Ra predominantly on the surface.
 - Nitric acid (1.8% HNO_3) did nothing.
 - Pickling (0.6% HF / 7.6% HNO_3) caused only 4.0 ± 1.6 x reduction.

Was acid etching of Ti IVC surface ineffective?

Test of AstroPak Etching of Ti

- LZ ICV etching performed by AstroPak
 - Avesta Red 240 Pickling Spray.
 - 45 minutes removed 14 μm .
- Recently performed test of emanation before/after AstroPak etching of 11 Ti discs.
 - 3850 cm^2 total.
 - For pre-etching measurements, performed emanations at ~ 100 mTorr and ~ 975 Torr.
 - After subtraction of blank runs and nuts used as spacers between discs, obtain
 - High-pressure: 0.77 ± 0.06 mBq
 - Diffusion and recoils
 - Low-pressure : 0.38 ± 0.06 mBq
 - Diffusion ~only
 - Suggests $\sim$ half at HP due to recoils.



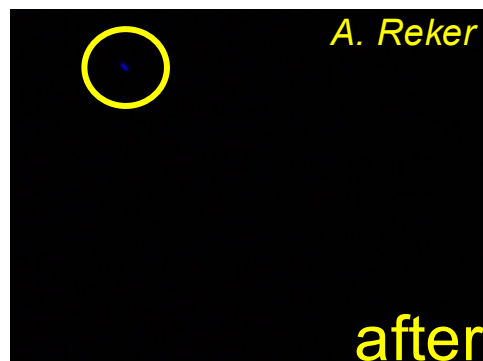
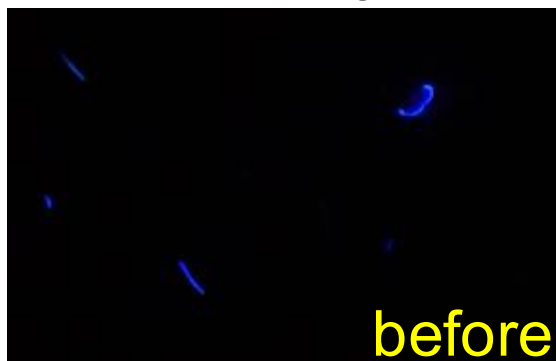
Results of (High-P) Post-etch Emanation

- AstroPak applied Avesta Red 240 Pickling Spray to the 11 Ti discs.
 - 150 μm removed from 6.
 - 11 μm removed from 5.
- Emanations performed at 975 Torr.
 - All 11 discs emanated together.
 - After subtractions of nuts and blanks, measured 0.15 ± 0.05 mBq (compared to original 0.77 ± 0.06 mBq).
 - PRELIMINARY, analysis and more data runs in progress
 - Implies reduction of factor 5.2 ± 1.8 .
 - Consistent with perfect removal at 3σ .
 - Consistent with XENON1T pickling of Ti.
 - If so, not as large as we would like.
- Wider XLZD Ti surface R&D includes Birmingham, Boulby, and Sheffield.

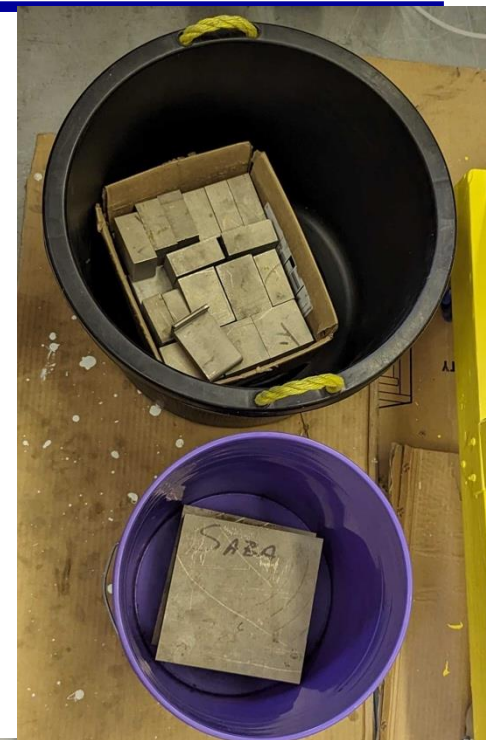


Titanium Blocks at SD Mines

- Ti cutouts from LZ assayed at SD Mines.
 - 31 pieces totaling 0.74 m², filthy when acquired.
 - Cleaned multiple ways:
 - Wiping with teflon brushes, wipes.
 - Ultrasonic cleaning.
 - Power washing (like LZ cryostat).
 - Dust assays using 44x camera under UV in dark space – enough magnification for most dust mass.
 - Power washing most effective, ~10x reduction.



- Preliminary radon emanation results ~5 mBq.
 - ~7x higher than etched LZ ICV.
 - Suggests etching reduced Ra ~7x for LZ ICV.



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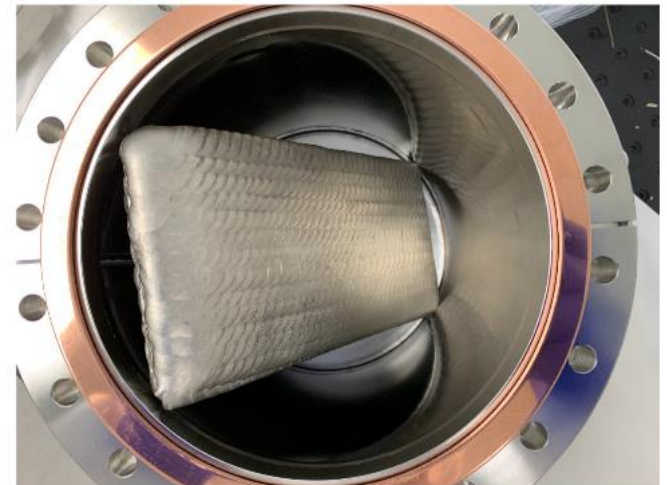
2) Radium concentrated near surfaces but surface etching not sufficiently effective?

3) Contamination added during assembly (e.g. welds)?

- Ti plate covered with 325 cm² TIG welds assayed at UCL.

- Used for QC via HPGe screening during assembly.
- 17.2 ± 3.0 mBq/m² untreated
- 11.4 ± 2.1 mBq/m² after etching
 - Scales to 4.9 ± 0.9 mBq in LZ, about 1/3 of the total.

U. Utku, Ph.D. thesis, University College London (2021).



Welds can be a significant problem.
Welds cannot be the main issue.

Conclusions & Plans for Future

- Titanium is attractive for low-background cryostats.
- High radon emanation from LZ Ti is becoming understood enough that future experiments should be able to avoid.
 - Expect large fraction of Ra contamination to be near surface.
 - May have significant contribution from welds.
 - Diffusion length < 0.2 mm. Lorenzo: Diffusion length negligibly short.
 - Acid etches attempted previously not sufficient.
 - (Still be careful not to add contamination during assembly!)
- XLZD and others performing R&D towards better solution.
 - XLZD overview in Ferdos Dastgiri's talk (T 15:35)
 - Electroplating -- see talks by Sophie Armbruster (M 13:50) and Margherita Noia (W 10:40)
 - Machining off surface material
 - Alternative acid etches
(Patrick Knights' poster)

*This work supported in part by US
Department of Energy (Grants No. DE-
SC0014223, DE-AC02-05CH11231)*