

Using Metal Organic Frameworks for Krypton and Radon Removal in Low-Background Xenon Detectors

Eric H. Miller^{1,*}, Brian Mong^{1,+}, Chamindu Amarasinghe², Maris Arthurs², Dongqing Huang², Wolfgang Lorenzon², Andrea Pocar³, and Richard Schnee⁴

¹SLAC National Accelerator Laboratory, Menlo Park, CA 94025, USA

²University of Michigan, Randall Laboratory of Physics, Ann Arbor, MI 48109, USA

³University of Massachusetts, Amherst, MA 01003, USA

⁴South Dakota School of Mines and Technology, Rapid City, SD 57701, USA

*emiller@slac.stanford.edu

+bung@slac.stanford.edu

August 31, 2020

Abstract

Here we express an interest to explore metal-organic-frameworks (MOF) as a means to separate the contaminants radon and krypton from xenon for future large xenon based detectors. MOF may also be a way to improve radon removal systems used to provide radon free clean spaces to build low background detectors.

Large xenon TPC experiments are used to search for new physics beyond the standard model such as dark matter interactions or neutrinoless double beta ($0\nu\beta\beta$) decay. Large detectors in the several ton to kilo-ton size are imagined to be employed in this search if current generation experiments do not find new physics. In these rare event searches, backgrounds from radioactive impurities present a problem for the identification of extremely rare events of interest. Due to the monolithic nature of the xenon-TPC, backgrounds originating from the detector materials are significantly suppressed by xenon's excellent self shielding ability. Backgrounds dissolved in the xenon material however, like the noble gases krypton and radon, are not reduced by self shielding and are not removed by the standard getter purifiers.

Krypton can get into the xenon supply during production and is found at the part per billion levels in typical research grade supply. Once removed however, krypton can only get back in to the xenon through air leaks during the experiment. Radon atoms, on the other hand, are emanated by the detector materials and gas handling system from the trace amounts of ^{238}U found in all materials, therefore these atoms need to be removed continuously during the experiment.

^{222}Rn is expected to be a major background source in the LZ (10,700kg) and nEXO (5,000 kg) experiments. In LZ the ^{222}Rn daughter ^{214}Po can produce untagged beta decays in the fiducial volume, and in nEXO the ^{214}Bi can emit a gamma ray with the same energy as the Q-value of $0\nu\beta\beta$ decay. LZ also requires krypton concentration at sub-PPT levels due to the ^{87}Kr ($T_{1/2} = 10.8$ yr) beta decay, which requires significant effort to achieve. As detectors continue to increase in sensitivity (and size) the background sources are increasingly dominated by xenon contaminants that are unaffected by the self shielding effect.

A number of noble-gas based experiments have used granulated activated charcoal (GAC) for the removal of gas contaminants. For example the XMASS collaboration has demonstrated the removal of radon from xenon in an inline GAC trap [1], since radon has a slower mean propagation velocity than xenon in the GAC trap, a trap could be designed to have the majority of radon decays ($T_{1/2} = 3.8$ days) occur before

passing through. The removal of krypton from xenon, as demonstrated by the LUX collaboration [2], is done in a similar fashion where krypton has a faster mean velocity in a GAC trap, and can be captured before the xenon reaches the end of the trap, and thus can be separated when dispensed in bursts of gas flow. The LZ collaboration is currently processing their entire xenon stockpile using GAC to remove krypton [3]. An existing alternative to GAC is distillation, which has been demonstrated by the Xenon collaboration for the removal of Krypton [4] and Radon [5] from xenon. There are challenges with scaling this technology, at least for online radon removal, to current and next generation experimental demands of 100's to 10,000's of SLPM flows. To achieve > 1000 SLPM flows, it is perhaps most feasible with liquid recirculation, and a liquid trap may be an energy and space efficient technique worth investigating.

There are two main drawbacks for using GAC traps: (1) Xenon binds strongly to charcoal, resulting in a high site occupancy fraction. In steady-state at 1 atm, a charcoal bed will adsorb its mass of xenon, which can become prohibitively expensive as charcoal masses exceed 100s of kg. (2) Table 1 shows radon emanation rates for various charcoals [6]. For ultrapure media, like the liquid xenon of a rare event search, a charcoal filter will eventually contribute more radon than it removes; this occurs for GAC traps capable of only a few 10's of SLPM gas flow.

Charcoal	Rn activity (mBq/kg)	Cost (USD/kg)
Calgon OVC 4x8	53.6 ± 1.3	6
Shirasagi G2x4/6-1	101.0 ± 8.0	27
Saratech	1.71 ± 0.20	35
HNO ₃ etched Saratech	0.51 ± 0.09	135
Carboact	0.23 ± 0.19	15,000

Table 1: ²²²Rn production rates and costs of various charcoals. Reproduced from [6].

Low background experiments have a unique need to create large clean spaces free from radon in air for detector construction and operation. This can be to reduce ²¹⁰Pb plateout on detector materials [7, 8] during construction, or to create an exclusion zone free from radon-daughter gammas in volumes around a running detector [9]. There are two current methods to remove radon from breathable air, both using GAC trap. One method is Vacuum Swing Absorption [7, 8] where air is flushed through one of two columns until the radon starts to breakthrough, then the column is switched. The non-flowing column is back-purged with some fraction of the radon reduced air under low pressure to regenerate the column. A second method passes air through a GAC column at cryogenic temperatures where the radon mean velocity is again slow enough that the radon decays in the column [10]. Both of these techniques have significant power and size requirements for cryogenic or vacuum pumps, and the large amounts of GAC (100's of kg). There now even exists a company, Ateko, that has built a number of Radon Removal Systems for these facilities [11].

Metal Organic Frameworks (MOF) are an interesting new development that may be a useful alternative for GAC in all of the applications mentioned above. MOFs are highly tunable materials [12] that can be designed to trap a particular species of gas, and are currently being investigated for xenon and krypton trapping [13], xenon-krypton separation [14], and radon removal from air [15]. There are even some investigations to recover xenon from air using MOF technology [16]. An MOF seems to be capable of 1000x more selectivity than GAC to the specific gas it is designed for, reducing the trap mass and size and, since MOFs are manufactured, it is possible that ingredients could be used that are low in ²³⁸U thereby removing the self-emanation problem.

References

- [1] K. Abe et al. Radon removal from gaseous xenon with activated charcoal. *Nucl. Instr. Meth. Phys. Res. A*, 661:50–57, 2012.
- [2] D.S. Akerib et al. Chromatographic separation of radioactive noble gases from xenon. *Astropart. Phys.*, 97:80–87, 2018.

- [3] B.J. Mount et al. LUX-ZEPLIN (LZ) Technical Design Report. 3 2017.
- [4] E. Aprile et al. Removing krypton from xenon by cryogenic distillation to the ppq level. *The European Physical Journal C*, 77:275.
- [5] E. Aprile et al. ^{222}Rn removal by cryogenic distillation in the XENON100 experiment. *The European Physical Journal C*, 77:358.
- [6] K. Pushkin et al. Study of radon reduction in gases for rare event search experiments. *Nucl. Instr. Meth. Phys. Res. A*, 903, 9 2018.
- [7] A Pocar. *Low background techniques and experimental challenges for Borexino and its nylon vessels*. PhD thesis, Princeton University, 2003.
- [8] J. Street, R. Bunker, E. H. Miller, R. W. Schnee, S. Snyder, and J. So. Radon mitigation for the SuperCDMS SNOLAB dark matter experiment. *AIP Conference Proceedings*, 1921(1):050002, 2018.
- [9] J. B. Albert et al. Search for neutrinoless double-beta decay with the upgraded EXO-200 detector. *Phys. Rev. Lett.*, 120:072701, Feb 2018.
- [10] A. Nachab. Radon reduction and radon monitoring in the NEMO experiment. *AIP Conference Proceedings*, 897(1):35–38, 2007.
- [11] Ateko. Available at https://www.ateko.cz/media/cache/file/25/ATEKO_Radon_Removal_Systems_2020.pdf (2020/08/26), 2020.
- [12] Avery E Baumann, D. A. Burns, Bingqian Liu, and V. S. Thoi. Metal-organic framework functionalization and design strategies for advanced electrochemical energy storage devices. *Communications Chemistry*, 2:1–14, 2019.
- [13] D. Bannerjee et al. Potential of metal–organic frameworks for separation of xenon and krypton. *Accounts of Chemical Research*, 48(2):211–219, 2015. PMID: 25479165.
- [14] A. Monpezad et al. Evaluation methods of adsorbents for air purification and gas separation at low concentration: Case studies on xenon and krypton. *Industrial Engineering Chemistry Research*, 58, 3 2019.
- [15] Xiaofei Zeng, Fei Chen, and Dapeng Cao. Screening Metal-Organic Frameworks for Capturing Radioactive Gas Rn in Indoor Air. *Journal of Hazardous Materials*, 366, 12 2018.
- [16] K. Elsaidi, S. et al. Xenon recovery at room temperature using metal–organic frameworks. *Chemistry – A European Journal*, 23(45):10758–10762, 2017.