Radon Burden and Xe Purification Performance of Hot Zirconium Getters

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(for the LUX-ZEPLIN collaboration)

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Abstract. We report on the radon emanation rate and xenon purification performance of hot zirconium getters, with a focus on their application to the LUX-ZEPLIN (LZ) dark matter experiment. New radon emanation measurements and HPGe gamma screening measurements of the getter pills and their precursor materials are presented, along with the performance of the 'Megatorr' purifier employed by LZ to remove electronegatives and other impurities from its Xe inventory.

INTRODUCTION

Entegris [1] noble gas purifiers are based upon St707 getter pill technology, a Zirconium (70%) – Vanadium (25%) – Iron (5%) alloy that chemically captures non-noble gas species on its surface [2]. The pills are heated to 400°C to promote the diffusion of captured impurities into the bulk, leaving the surface free for additional gettering. St707 is effective at removing electronegatives such as O_2 and H_2O, as well as N_2, CH_4, and other species. For this reason these purifiers have been widely adopted by xenon TPC experiments, including the LUX-ZEPLIN (LZ) dark matter experiment [3, 4]. Prior experience has found that for each tonne of Xe to be purified, roughly 40 to 80 SLPM of continuously recirculating gas flow is required to maintain an acceptable free electron lifetime. High flow rates require efficient gas heat exchange in the purifier, and a large mass of getter pills. However, for low background experiments, the pills present a modest but non-negligible radon burden which can contaminate the experiment with alpha, beta, and gamma decays.

We studied the radon emanation rate of these getters, along with their purification performance, to gain insight on how to mitigate and reduce the radon burden. Our findings suggest two simple but effective strategies: screen and select a favorable lot of getter pills; and choose the smallest purifier model that satisfies the purification requirements of the experiment.

RN EMANATION RATE OF THE ENTEGRIS PS4-MT50

The Entegris PS4-MT50 purifer contains 4.4 kg of St707 getter pills. This model is recommended by the manufacturer for Xe gas flow rates up to 100 SLPM. We performed ^{222}Rn emanation measurements of the PS4-MT50 at the University of Maryland with an electrostatic counter (ESC). Helium is used as a carrier gas to transport the Rn from the purifier to the counter. The helium is prepared by passing it through a carbon trap at 77 K to remove latent radon. The PS4-MT50 is purged with the clean helium several times, filled to a pressure of one atmosphere, and sealed. After emanating radon for one to two weeks, the purifier is again purged with helium, and the exhaust gas stream is passed through a copper bead trap at 77 K where the radon atoms attach to the bead surfaces. The helium is evacuated from the trap. When the trap returns to room temperature, the ^{222}Rn atoms are released from the copper and are flushed into the 1.7-liter ESC by volume-sharing followed by another clean helium stream. (Carbon is not used for this trap because it does not efficiently release radon at room temperature.) The total transfer efficiency is close to 100%, as confirmed by calibration measurements. ^{222}Rn decays in the ESC create positively charged ^{218}Po ions, which are collected with an electric field onto an Ortec Ultra-AS surface barrier diode. The counter efficiency is 24% based upon a comparison to a calibration standard. Its inefficiency is dominated by the alpha decay acceptance of the diode (50%) and the ^{218}Po ion fraction (59%) [5]. The system is depicted in Figure 1.

Initial measurements indicated that the radon emanation rate of the particular unit under study was close to the limit of detection of the system, which is ~ 0.15 mBq. To gain precision, six measurements were made, three with the

FIGURE 1. Left: St707 getter pills harvested from a used PS4-MT3 noble gas purifier. The darker pills in the lower right are oxidized from prior use. **Center:** The Entegris PS4-MT50 purifier. The getter pill vessel is inside the grey heater blanket on the right. **Right:** Radon transfer and counting system at the University of Maryland. Charcoal and copper traps are used to prepare clean helium and to collect and transfer the radon. The 1.7-liter ESC is the conflat vessel at lower right.

TABLE I. Radon emanation measurements of Entegris noble gas purifiers. Results are reported in terms of the equilibrium decay rate. The Maryland results are a likelihood average of three measurements for each temperature. Assuming that the St707 getter pills are the radon source, the specific activity is computed by dividing by the pill mass (4.4 kg for the PS4-MT50, and 15 kg for the PS5-MGT50). The Xenon1T measurements refer to items 16 and 17 in Ref. [6]

Measurement	Model	Temperature	^{222}Rn (μBq)	^{222}Rn Specific Activity (μBq/kg)
Maryland	PS4-MT50	ambient	80 ± 30	18 ± 7
Xenon1T (ID #16)	PS4-MT50	ambient	610 ± 40	138 ± 9
Maryland	PS4-MT50	400 C	142 ± 37	32 ± 8
Xenon1T (ID #16)	PS4-MT50	400 C	1170 ± 150	265 ± 38
Xenon1T (ID #17)	PS4-MT50	400 C	240 ± 30	55 ± 7
LZ [7]	PS5-MGT50	400 C	2260 ± 270	151 ± 18

purifier emanating at room temperature, and three at 400 °C. The measurements at each temperature are consistent with each other and are combined with a likelihood analysis.

The results are shown in Table I, along with measurements performed on the same purifier model by the Xenon1T collaboration [6]. A comparison of the three units shows a factor of seven variation in the radon emanation rate. The measurements also indicate that the emanation rate increases when the purifier is heated to its operating temperature, consistent with the assumption that the pills are the primary source of radon. Taken together, the results suggest that a careful selection of the pills could be beneficial.

HPGE GAMMA SCREENING MEASUREMENTS OF ST707 GETTER PILLS

To further investigate the origin of the radon activity, HPGe gamma screening measurements were performed on samples of SAES St707 getter pills and its precursor materials (the raw ingredients). The goal was to detect the gamma lines below ^{226}Ra, and compare the inferred activity to the ^{222}Rn emanation rate attributed to the pills.

The results of measurements performed at the Berkeley Low Background Facility are shown in Table II and Table III. The pills were found to contain a substantial amount of actinium chain activity. The ^{238}U and ^{232}Th chains were also detected. All three chains are out of secular equilibrium. The actinium chain is observed for ^{231}Pa and below at a level of 10 – 15 Bq/kg. The measurements of the precursor materials provided by SAES identified the zirconium as the source of ^{231}Pa and its daughters. A second sample of zirconium, obtained independently from Loterios [8], also exhibited the same pattern of actinium-chain activity. The presence of the thorium and actinium chains limited the sensitivity of the measurements to the ^{226}Ra chain. In any case, neither this zirconium nor the getter pills can be considered to be low-background materials. Finding a cleaner source of zirconium may enable the fabrication of

TABLE II. ^{238}U, ^{232}Th, and ^{40}K activity of SAES getter pills and precursor materials. Measured at the Berkeley Low Background Facility with an HPGe detector. ^{238}U (late) refers to ^{226}Ra and its daughters.

Material	Comment	^{238}U (early) (mBq/kg)	^{238}U (late) (mBq/kg)	^{232}Th (early) (mBq/kg)	^{232}Th (late) (mBq/kg)	^{40}K (mBq/kg)
SAES St707 pills	used	2600	< 10	130	70	< 80
SAES St707 pills	new	11000	< 50	20	30	220
SAES Zr #1		480	< 40	< 55	< 20	160
Loterios Zr #2		< 250	< 40	72	< 20	900
SAES V-Fe alloy		< 170	< 30	< 40	< 20	160

TABLE III. Actinium chain activity of SAES getter pills and precursor materials. Measured at the Berkeley Low Background Facility with an HPGe detector.

Material	Comment	^{235}U (mBq/kg)	^{231}Pa (mBq/kg)	^{227}Th (mBq/kg)	^{223}Ra (mBq/kg)	^{219}Rn (mBq/kg)	^{211}Pb (mBq/kg)
SAES St707 pills	used	90	14700	6900	7500	7200	7600
SAES St707 pills	new	600	16700	2800	3400	3300	3500
SAES Zr #1		90	15100	2300	2700	2200	2500
Loterios Zr #2		90	10500	1000	870	750	900
SAES Fe-V alloy		< 16	< 120	–	–	–	–

improved getter pills.

PERFORMANCE OF THE LZ MEGATORR PURIFIER

The LUX-ZEPLIN (LZ) experiment is a large liquid xenon TPC searching for scatters of WIMP dark matter. It is currently collecting data at the Sanford Underground Research Facility (SURF) in Lead, SD. LZ employs an Entegris 'Megatorr' PS5-MGT50 purifier containing 15 kg of getter pills to remove electronegatives from its 10.4-tonne Xe inventory. Its observed emanation rate is 2.26 ± 0.27 mBq [7]. If we attribute this radon to the getter pills, the specific activity is about five times higher than that of the Maryland PS4-MT50 (see Table I). The two units do not share getter pills from the same lot, so this may be an example of production variation.

The design flow rate of the LZ purification system is 500 SLPM, and Entegris recommends a Megatorr model known as the PS5-MGT100 for Xe gas flows between 275–550 SLPM. LZ elected to deploy the half-size MGT50 mainly to reduce the expected radon by a factor of two. The LZ plumbing system is designed to allow a second MGT50 unit to be connected in parallel if the purification performance of the system is poor. However, electron lifetimes in excess of 7 msec have been achieved in LZ [4], and the overall purifier performance has been satisfactory.

The temperature of the getter bed is a key parameter which determines the purifier's one-pass removal efficiency, and Entegris provides integrated thermometry to monitor it. The Megatorr also has a gas pre-heater and gas-gas heat exchanger to reduce the thermal burden of the flowing gas on the getter pills. Ideally, the gas should be heated to the operating temperature of 400°C before encountering the getter bed, to prevent the pills from being cooled.

As shown in Figure 2, the getter bed temperature of the LZ Megatorr initially dropped from the nominal 405°C to 365°C during Xe gas flow startup; however the temperature recovered to > 400°C within five hours while accommodating a gas flow rate of 500 SLPM. In a later test, the gas flow rate was increased to 600 SLPM without negatively impacting the vessel bed temperature. Another important design parameter of the Xe circulation system is the pressure drop as a function of gas flow rate; this is shown for the LZ system in Figure 2.

Methane is one of the most difficult species to remove from Xe due to its highly inert nature [9]. The Megatorr purifier has a methane capacity of 20 grams. As a test of purifier performance, 0.1 standard liters (0.07 g) of natural methane (CH_4) was injected into the 10.4-tonne Xe inventory in October 2021. The methane concentration in the detector was observed for the next three weeks with a cold trap mass spectrometry system [10, 11]. The results are shown in Figure 3. The initial concentration was found to be 4.1 ppb (g/g), roughly consistent with the injected quantity. As the purification system processed the Xe, the methane concentration dropped exponentially with a time constant of $\tau = 3.58 \pm 0.08$ days. The Xe gas flow rate during this period varied from 390 to 600 SLPM, with an

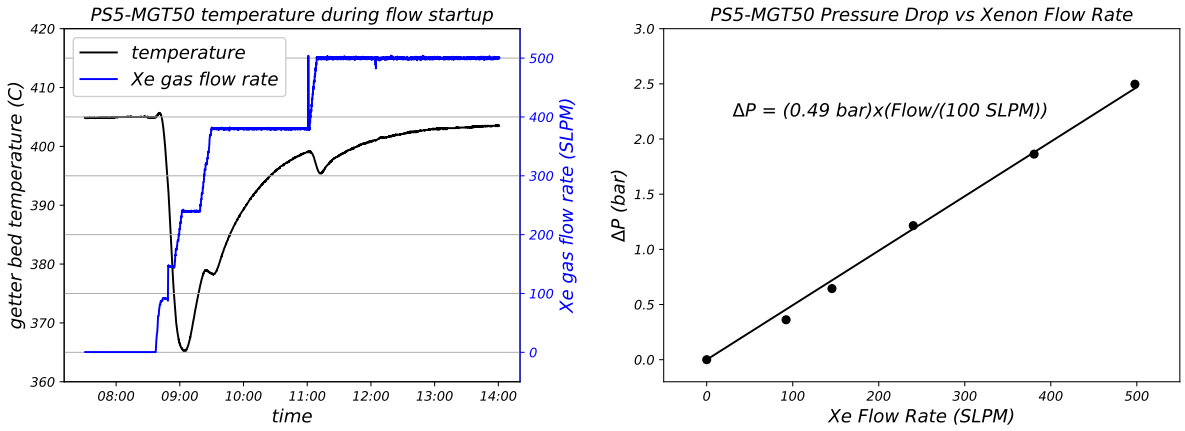

FIGURE 2. Left: Temperature of the LZ Megatorr purifier during Xe gas flow startup. Black: getter bed temperature (left axis); blue: Xe gas flow rate (right axis). The vessel temperature starts at 405°C with zero gas flow. As the gas flow ramps up to 500 SLPM, the getter temperaure drops to 365°C, but eventually recovers to > 400°C. **Right:** The pressure drop across the purifier as a function of Xe gas flow rate.

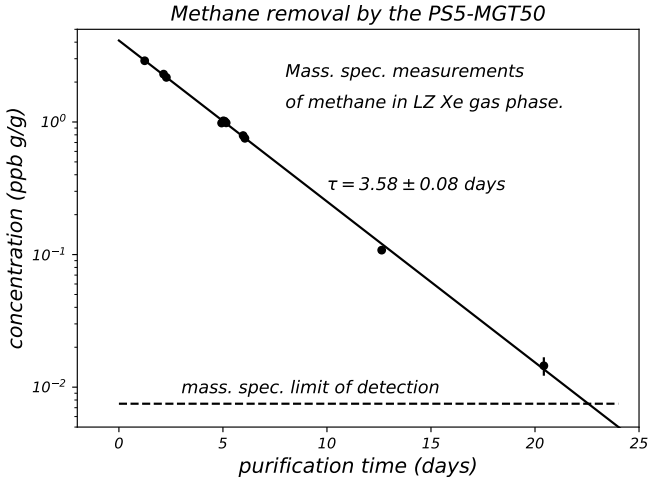

FIGURE 3. Removal of methane from Xe by the LZ Megatorr purifier. The methane concentration is measured by a cold trap mass spectrometry system. The observed concentration is fitted to a decaying exponential: $\rho(t) = \rho_0 * exp(-t/\tau)$.

average value of 516 SLPM. This flow rate implies that a removal time constant of 2.4 days should be expected, somewhat less than the observed value. In fact, at the time of the test, the Megatorr temperature control system was holding the getter bed at 320°C, 80 degrees below the intended set point. This issue was later corrected, and future measurements are planned to determine if the methane removal time has been impacted.

CONCLUSION

Several lessons can be discerned regarding the minimization of the radon burden of hot zirconium getters. First, the lot-to-lot variation in the radon emanation rate of the getter pills appears to be non-negligible. Careful selection of the pills may allow for further improvement. To screen the pills, HPGe gamma detection is a potential alternative to direct radon counting, however this method can be inconclusive if there is substantial thorium and actinium chain activity. Cleaner sources of zirconium could improve this situation, and may allow cleaner getter pills to be manufactured.

Finally, a crude but effective strategy for xenon-based experiments is to choose a smaller purifier model than the one recommended by Entegris.

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