RADON BURDEN AND XENON PURIFICATION PERFORMANCE OF HOT ZIRCONIUM GETTERS

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INTRODUCTION

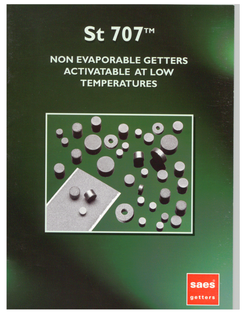
ENTEGRIS (formerly SAES PURE GAS) noble gas purifiers are loaded with SAES St707 getter pills, a Zirconium(70%)-Vanadium(25%)-Iron(5%) alloy that chemically captures non-noble gas species on its surface. The pills are heated to 400 C to promote diffusion of captured impurities into the bulk, leaving the surface free for additional gettering.

Xenon TPC experiments must typically achieve an electron lifetime greater than one millisecond to function properly, and ENTEGRIS purifiers are a common solution. For each tonne of Xe to be purified, 40 to 80 SLPM of flow is typically required to maintain acceptable purity. High flow rates require efficient heat exchange, and therefore a large mass of getter pills; however, the pills present an unwanted radon burden for low background experiments.

We studied the radon emanation of these purifiers to gain insight on how to mitigate and reduce it for low background Xe experiments. Our findings suggest two simple strategies: (1) select a favorable lot of getter pills; (2) choose the smallest purifier model which meets the heat exchange requirements of the Xe gas flow conditions.

HIGHLIGHTS

- Entegris hot zirconium purifiers can reduce electronegative contaminants in Xe to concentrations below 0.1 ppb (O_2 equivalent), corresponding to an electron lifetime in liquid xenon greater than 3 milliseconds.
- The ^{222}Rn emanation rate of the purifiers increases when the purifier is at its operational temperature of 400 C. This suggests that the getter pills are the source of the radon.
- The specific activity of the hot getter pills varies between 32 and 265 μBq/kg in the same purifier model (PS4-MT50).
- HPGe gamma screening of the getter pills finds significant activity in ^{238}U, ^{232}Th, and ^{235}U (Actinium) chains. Secular equilibrium is broken for all three chains.
- The most prominent activity is in the Actinium chain below ^{231}Pa, which is present at the level 10 – 15 Bq/kg.
- A HPGe analysis of getter pill precursors found that the Actinium chain activity is introduced by the Zirconium raw material. Zirconium sourced from an independent supplier had the same contamination.
- The high level of Actinium chain activity obscures the observation of gamma lines from ^{226}Ra, the parent of ^{222}Rn. Limits on ^{226}Ra are in the range of 10's of mBq/kg.
- The LZ experiment employs a large Entegris Megatorr purifier, model PS5-MGT50-R-535. The ^{222}Rn emanation rate is 2.26 ± 0.27 mBq.
- Removal of methane and other impurity species by the LZ purifier has been observed with a mass spectrometry system. The electron lifetime is 7 milliseconds.
- Entegris/SAES recommends a Xe gas flow rate < 275 SLPM for this model purifier, but LZ has achieved good performance at 500 SLPM while maintaining a getter vessel bed temperature of 400 C. The pressure drop across the purifier is 2.5 bar at 500 SLPM.



Left: St707 getter pills harvested from a SAES PS4-MT3 noble gas purifier. The shiny pills in the upper left are mostly unused; the darker pills in the lower right are oxidized from prior use. Right: SAES getter pill promotional brochure.

^{222}RN EMANATION RATE OF THE PS4-MT50

^{222}Rn emanation measurements of PS4-MT50 units have been measured at the University of Maryland. The University of Maryland operates an electrostatic Rn counter originally constructed by the Shutt group at Case Western Reserve University. The efficiency of the counter is 24% based upon comparison to a calibration standard. An ion drift simulation is shown in the inset below.

Left: The PS4-MT50 purifier at the University of Maryland. The getter vessel is inside the grey heater blanket on the right side. Right: Maryland Rn counting apparatus. Charcoal and copper traps are used to transfer Rn from the getter to the electrostatics counter (Conflat vessel at lower right). A silicon barrier detector (Ortec) is used to count ^{218}Po ^{214}Po, and other Rn daughters. Inset: A simulation of ion drift in the counter.

The Maryland PS4-MT50 has a radon emanation rate is close to the limit of detection of the counting system. To gain precision, six measurements were made, three at room temperature, and three at 400 C. The measurements were then combined with a likelihood analysis. Two purifiers of the same model were also measured Xenon1T collaboration[2]. A comparison of the three units shows a Rn emanation variation of a factor of seven.

SAES Monotorr PS4-MT50 Rn emanation measurements

Measurement	Temperature	Activity (μBq)	Getter Pill Mass (kg)	Activity (μBq/kg)
Maryland 1	room temp	79 + 55 - 42		
Maryland 2	room temp	133 + 66 -54		
Maryland 3	room temp	1 + 57 - 44		
Maryland Combined	**room temp**	**80 + 33 - 28**	**4.4**	**18 ± 7**
Xenon1T (ID #16)[2]	**room temp**	**610 ± 40**	**4.4**	**138 ± 9**
Maryland 4	400 C	121 + 71 - 56		
Maryland 5	400 C	168 + 80 - 68		
Maryland 6	400 C	119 + 58 - 46		
Maryland Combined	**400 C**	**142 + 39 - 35**	**4.4**	**32±8**
Xenon1T (ID #16)[2]	**400 C**	**1170 ± 150**	**4.4**	**265 ± 38**
Xenon1T (ID #17)[2]	**400 C**	**240 ± 30**	**4.4**	**55 ± 7**

HPGE STUDIES OF GETTER PILL ACTIVITY

HPGe gamma screening measurements were performed on samples of getter pills and getter pill precursor materials. The measurements were performed at the Berkeley Low Background Facility at Lawrence Berkeley National Lab.

The pills were found to be contaminated with an unusually high quantity of Actinium chain (235U) activity. The 238U and 232Th chains were also detected. All three chains are out of secular equilibrium, with the Actinium chain being observed below ^{231}Pa at a level of 10-15 Bq/kg. The precursor measurements identified the Zirconium metal as the source of the Actinium chain. A second sample of Zirconium, obtained from Loterios, also exhibited the same Actinium chain activity below ^{231}Pa. The presence of the Actinium chain limited the sensitivity of the measurements to the ^{226}Ra (^{238}U(late)) parent of ^{222}Rn.

HPGe gamma screening of getter pills & precursors – Uranium, Thorium, and Potassium chains

Material	Comment	^{238}U(early)	^{238}U(late)	^{232}Th(early)	^{232}Th(late)	^{40}K
		(mBq/kg)	(mBq/kg)	(mBq/kg)	(mBq/kg)	(mBq/kg)
SAES St707 getter pills	used	2600	< 10	130	70	< 80
SAES St707 getter pills	new	11000	< 50	20	30	220
Zirconium #1 (getter pill precursor)	From SAES	481	< 37	< 53	< 24	155
Zirconium #2	From Loterios	< 247	< 37	73	< 20	898
V-Fe Alloy (getter pill precursor)	From SAES	< 173	< 37	<41	< 20	164

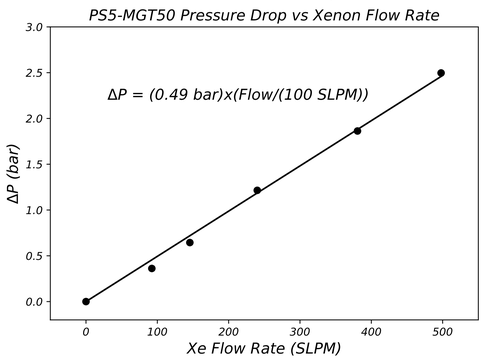
HPGe gamma screening of getter pills & precursors - Actinum chain

		^{235}U	^{231}Pa	^{227}Th	^{223}Ra	^{219}Rn	^{211}Pb
		(mBq/kg)	(mBq/kg)	(mBq/kg)	(mBq/kg)	(mBq/kg)	(mBq/kg)
SAES St707 getter pills	used	90	14700	6900	7500	7200	7600
SAES St707 getter pills	new	600	16700	2800	3400	3300	3500
Zirconium #1 (getter pill precursor)	From SAES	90	15100	2300	2700	2200	2500
Zirconium #2	From Loterios	90	10500	1000	870	0750	900
Fe-V Alloy (getter pill precursor)	From SAES	< 16	< 120	-	-	-	-

PERFORMANCE OF THE LZ PS5-MGT50

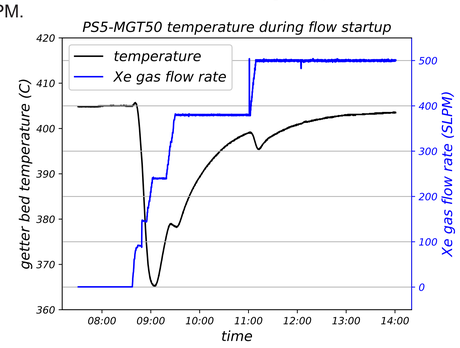
LZ employs an Entegris PS5-MGT-R-535 purifier to remove electronegatives from its 10 tonne Xe inventory. The Rn burden of this unit is 2.26 ± 0.27 mBq [1]. The mass of getter pills is not known; however, it is likely to be between 10 and 20 kg, giving an approximate specific activity of 110 – 230 μBq/kg. This is in the upper half of the range observed for the PS4-MT50 units. This suggests that it may be possible to identify cleaner getter pills that would reduce the Rn burden of the PS5-MGT50 by a factor of three to six.

LZ has achieved Xe flow rates up to 500 SLPM through the PS5-MGT50, although Entegris recommends a larger unit (the PS5-MGT100) for flows greater than 275 SLPM. The pressure drop across the purifier as a function of Xe flow rate is shown below. At 500 SLPM, the ΔP is 2.5 bar.

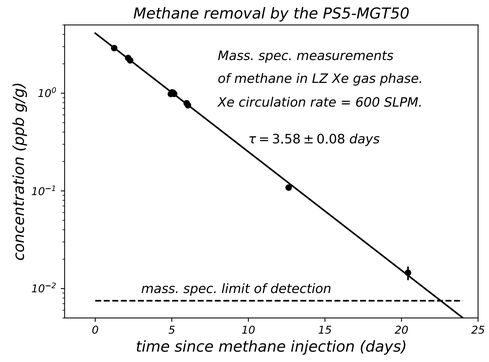


The PS5-MGT50 purifier from Entegris.

For flow rates exceeding the manufacturer's recommendation, cooling of the getter pills by the Xe gas is a concern. The PS5-MGT50 getter bed has an integrated thermometer allowing observation of its temperature. During a flow ramp-up event, shown below, the getter bed temperature experienced a temporary temperature drop from 400 to 365 C, but the temperature soon recovered, indicating adequate heat exchange at 500 SLPM.



Removal of methane is a stringent test of purifier performance. After initiating Xe circulation in LZ, an experiment was performed where methane was injected into the Xe inventory at a concentration of 30 ppb. The methane concentration was observed with a mass spectrometry system for the next month while circulating Xe through the PS5-MGT50. The results are shown below. At a flow rate of 600 SLPM, the removal time constant was found to be 3.58 ± 0.08 days, somewhat longer than the two-day Xe-turnover time expected at this flow rate. Due to a purifier control system error, the getter temperature was 310 C during this experiment, below the nominal value of 400 C. Future measurements are planned to determine if a normal or elevated getter temperature can produce a shorter methane removal time. Mass spec. measurements show that the getter is removing N_2 and other species very well, and the free electron lifetime in the TPC is 7 milliseconds, indicating good electronegative purity. The purification performance of the getter has been acceptable.



REFERENCES

[1] D.S. Akerib et al., EPJ C, Vol. 80: 1044 (2020), arXiv:2006.02506

[2] E. Aprile, et al., EPJ C, Vol. 81: 337 (2021), arXiv:2009.13981.

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