TREATMENT OF MERCURY TARGET OFF-GAS AT SNS

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The Spallation Neutron Source (SNS) is the first operational spallation source to use liquid Mercury as a target material. This paper describes the treatment system to remove volatile spallation products from a Helium purge stream that emanates from the Mercury target and adjustments made to achieve design goals in response to phenomena experienced during initial operations.

The Helium stream is treated to remove volatile spallation products prior to environmental release because of its activity level as these accumulate in the gas space in the Mercury Loop. Unanticipated local dose rates were noted in treatment system components during low power startup. Gamma scanning of these components identified the presence of nineteen noble gas isotopes and their daughters, indicating that the doses resulted from noble gas sorption. Significant moisture was also encountered in the system, resulting in the plugging of the system cold trap. Necessary re-configuration of Mercury pump components presented additional requirements.

The Off-Gas Treatment System has been successfully operated since April, 2006. System availability and removal effectiveness have been high. Operational issues occurring during the first year of operation have been resolved.

I. INTRODUCTION

The Spallation Neutron Source (SNS) is the first operational spallation source to use liquid Mercury as a target material[1]. This innovation has necessitated a different set of design choices not previously required in the operation of this type of facility. This paper describes one such issue regarding the production and control of volatile spallation products.

I.A. Production of Volatile Spallation Products from Hg

The spallation products vary in abundance from Hydrogen isotopes, which are relatively abundant, to other much less abundant species such as Cesium. As a general rule, except for Hydrogens and Heliums, the lower a spallation product's atomic number, the lower its relative abundance from Mercury spallation[2]. Spallation product gases that are predicted[3] to be created in the Mercury Target Loop are listed in Table I on the following page. These gases result both from direct spallation as well as from decay from parent isotopes.

Spallation products are created as gases (or are created from other spallation product isotopes that have gaseous daughter products in their decay chains) are essentially insoluble in mercury. In the SNS Mercury Target System, these gases accumulate in the head space at the top of the mercury pump tank, which is the highest point in the Mercury Loop.

Xenon isotopes ^{119}Xe to ^{125}Xe are of particular interest because they decay to Iodine species (Table II on the following page).

A more complete discussion of the creation of spallation products from proton reactions with Mercury may be found in Reference 2.

I. B. Mercury Offgas Treatment System Design

Because of operational and cost considerations, it was desirable to use a system that is passive to avoid frequent operator attention. The SNS Mercury Off-Gas Treatment System (MOTS) uses a combination of decay and sorption for treatment of spallation product gases. A low flow stream of helium originates from loop instrumentation and from the shaft seal purge on the mercury pump. This stream was also intended to purge the pump tank at a low flowrate which causes the pump tank to act as a decay tank due to the relatively long residence time. Helium was chosen because it is inert and non condensable at Liquid Nitrogen temperatures, and does not sorb on charcoal at operating temperatures.

Table I. Predicted Spallation Product Gases Produced from Mercury Spallation with 1 GeV protons

Isotope	Half Life	Isotope	Half Life	Isotope	Half Life	Isotope	Half Life
H	stable	^{37}Ar	35.0 d	^{83m}Kr	1.86 h	^{125}Xe	17.1 h
D	stable	^{38}Ar	stable	^{84}Kr	stable	^{126}Xe	stable
T	12.3 y	^{39}Ar	259 y	^{85}Kr	10.76 y	^{127}Xe	36.4 d
^{3}He	stable	^{40}Ar	stable	^{85m}Kr	4.48 h	^{128}Xe	stable
^{4}He	stable	^{41}Ar	1.827 h	^{86}Kr	stable	^{129}Xe	stable
^{13}N	9.97 m	^{42}Ar	33 y	^{87}Kr	1.27 h	^{130}Xe	stable
^{14}N	stable	^{43}Ar	5.4 m	^{88}Kr	2.84 h	^{131}Xe	stable
^{15}N	stable	^{44}Ar	11.87 m	^{89}Kr	3.15 m	^{131m}Xe	11.9 d
^{16}N	7.13 s	^{72}Kr	17 s	^{91}Kr	8.6 s	^{132}Xe	stable
^{14}O	70.6	^{73}Kr	27 s	^{92}Kr	1.29 s	^{133}Xe	5.243 d
^{15}O	122.2 s	^{74}Kr	11.5 m	^{93}Kr	1.84 s	^{133m}Xe	2.19 d
^{16}O	stable	^{75}Kr	4.3 m	^{94}Kr	1.29 s	^{134}Xe	stable
^{17}O	stable	^{76}Kr	14.8 h	^{95}Kr	0.78 s	^{135}Xe	9.1 h
^{18}O	stable	^{77}Kr	1.24 h	^{97}Kr	0.1 s	^{135m}Xe	15.3 m
^{18}Ne	stable	^{78}Kr	stable	^{119}Xe	5.8 m	^{137}Xe	3.82 m
^{19}Ne	17.22 s	^{79}Kr	1.455 d	^{120}Xe	40 m	^{138}Xe	14.1 m
^{20}Ne	stable	^{80}Kr	stable	^{121}Xe	39 m	^{139}Xe	39.7 s
^{21}Ne	stable	^{81}Kr	2.3E5 y	^{122}Xe	20.8 h	^{140}Xe	13.6 s
^{22}Ne	stable	^{82}Kr	stable	^{123}Xe	2 h	^{141}Xe	1.72 s
^{25}Ne	0.61 s	^{83}Kr	stable	^{124}Xe	stable	^{142}Xe	1.22 s
^{36}Ar	1.77 s						

Table II. Xenon Isotopes that have Iodine Daughters

Isotope	Daughter	Half Life
^{119}Xe	^{119}I	19.1 m
^{120}Xe	^{120}I	1.35 h
^{121}Xe	^{121}I	2.12 h
^{122}Xe	^{122}I	3.6 m
^{123}Xe	^{123}I	13.2 h
^{125}Xe	^{125}I	59.4 d

Because of the elevated mercury temperature in the loop, mercury vapor is also present in the helium stream. To confine the mercury to the service bay, the He offgas stream flows thru Gold Bed 1 where the mercury sorbs on the gold-on-alumina media. This innovative material is produced by ADA Technologies[4].

The He flow then continues out of the Service bay to the basement treatment area and thru a second Gold Bed unit, before entering the CuO reactor. Here, the CuO is reduced to metallic Cu and the hydrogen isotopes, particularly tritium, are converted to water vapor. The helium stream then flows through molecular sieve where the water vapor is removed. The use of CuO allows the conversion the hydrogen to water without the addition of oxygen to the system. The CuO reactor must be periodically regenerated with oxygen. The gas finally flows through a liquid nitrogen cooled charcoal adsorber which is operated at 77 K to remove noble gases and other residual activity. A flowsheet of the system is shown in Figure 1.

I. C. System Operational Experience

Beginning with the first beam-on-target in April, 2006 the system has been operating for all neutron production operations. Successful operation of the system is demonstrated by the lack of significant activity detected by the SNS stack monitor during beam-on-target operations. Several other operational data points, some of them unexpected, have been observed. During initial operations, at a beam power level of only 5 kW, a contact dose rate of approximately 0.7 mSv/h was observed at the second gold bed in the system, physically located in the basement treatment room. (Fig 1). A gamma scan of the system was conducted to identify isotopes. The results of this scan is shown in Figure 2, and resulted in the identification of nineteen noble gas isotopes and daughters in the system. These isotopes are listed in Table 3 below.

Generally, only isotopes with half lives longer that approximately 2 hours were identified. This is thought to result from a combination of factors, including the residence time on the mercury pump tank, the beam power, and the relatively short operating times. The latter results in non-equilibrium production of longer lived spallation product gases. The flowrate of helium from the loop is approximately 1 L/m (2 scfh) and the volume of

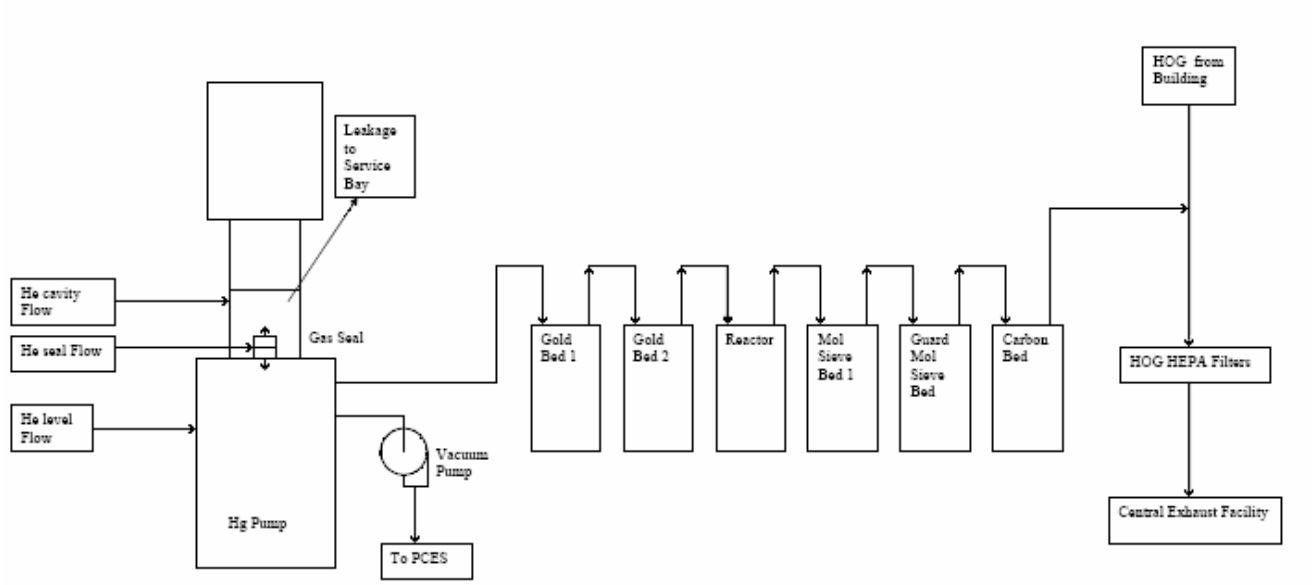

Fig. 1. MOTS Flowsheet

the pump tank free space during operation is approximately 0.11 m^3. This gives a residence time of approximately 2 hours. Short lived volatile gas species therefore are believed to decay in the mercury loop prior to reaching the basement treatment system area.

The cause for this buildup on the Gold unit determined to be the sorption of noble gases on the gold media. A measured adsorption coefficient for noble gases on gold has not been reported in the literature, but is thought to be several orders of magnitude below that of carbon at room temperature. However, due to the energetic nature of radiation from the positron emitting noble gases, very small quantities of gases can cause measurable dose rates.

During our initial low power operations, apparently all of the noble gases were being sorbed by the gold beds before they reached the carbon adsorber. We found that by treating the gold beds with stable xenon allowed the stable xenon to fill up the available sorption sites on the gold media thus drastically reducing sorption of radioactive xenons during operations. Treatment of the gold beds prior to installation in the system has prevented recurrence. Although these dose rates have been reduced, because noble gases are still transiting the bed, a dose rate of approximately 90 μSv/h contact persists at power levels of ~30 kW. Temporary shielding has been installed, but with the projected dose rates for higher proton beam power which are expected by 2009, may warrant permanent shielding.

In addition to sorption of noble gases, the gold media has had other issues. During early operations it was determined that this media sorbs moisture from the air. Since the last step in the media production procedure is heating to 400C, moisture driven off during manufacturing is re-adsorbed during shipment and storage of the media. The gold media was originally designed for removal of mercury from coal power plant flue gas; having moisture present for this application is not an issue. However in the MOTS application the influent gas is already very dry and apparently caused the sorbent to releases its moisture during operation. This shortened molecular sieve bed lifetime. During initial operations, the problem was compounded by the fact that mercury loop testing was initially done with water. Although the loop was dried by purging with compressed air prior to mercury filling, residual moisture from the mercury loop was also purged into the gold beds during the first loop trials.

The gold media therefore required early replacement. The new gold media was dried prior to use by heating and purging with dry nitrogen to a -30C dew point prior to installation. Subsequent to removal, the removed gold media was also dried using the same procedure, and approximately 1 liter of water was recovered. This water was found to contain tritium.

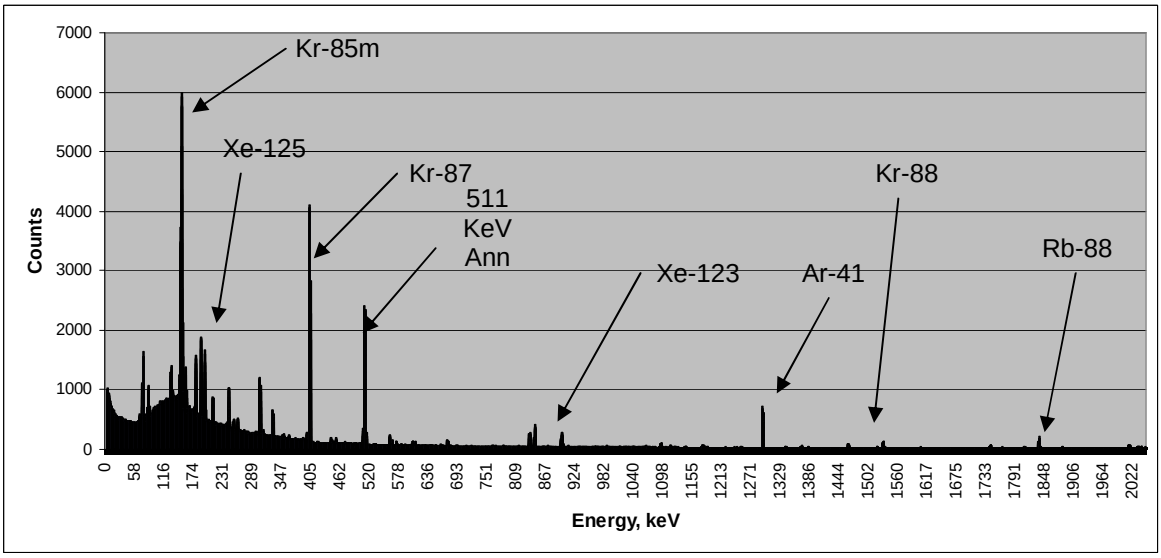

Fig. 2. SNS MOTS Gamma Spectrum, August, 2006

Table III Identified Spallation Product Gases (and their daughters) August, 2006

Isotope	Half Life	Daughter Isotope	Half Life	Daughter Isotope	Half Life
Ar-41	1.827 h				
		Br-76	16.2		
Kr-77	1.24 h	Br-77	57.04 h		
Kr-79	35.04 h				
Kr-85m	4.48 h				
Kr-87	76.3 m				
Kr-88	2.84 h	Rb-88	17.8 m		
Xe-120	40 m	I-120	81 m		
Xe-121	40.1 m	I-121	2.12 h	Te-121	16.8 d
Xe-122	20.1 h	I-122	3.63 h		
Xe-123	2 h	I-123	13.27 h		
Xe-125	16.9 h				
Xe-127	36.4 d				

During early operations this moisture also created a freeze plug in the cryogenic carbon bed. This occurred primarily as a result of operating the system with the dew point sensor reading of 20C in the outlet gas. The freeze plug was removed by heating, and the molecular sieve beds changed, but there was so much residual moisture in the system from the above mentioned sources that a molecular sieve bed only functioned for 8 hours before breakthrough was detected. The molecular sieve beds were therefore determined to be too small since they required these frequent changes. These beds were designed to remove moisture resulting from oxidation of spallation hydrogen, and not atmospheric moisture. Over time this situation improved to the order of 3 days time between changes, but the decision was made to install larger beds. During the July 2006 outage a different type of molecular sieve bed was installed. This new design worked well and therefore the remaining molecular sieve beds were replaced during the December 2006 outage. This design allows regeneration by heating, thus greatly reducing the need to replace the molecular sieve. No

molecular sieve has been replaced since the new beds have been installed.

II. SUMMARY AND CONCLUSION

The offgas treatment system for the helium purge stream from the SNS Target Mercury Loop has been successfully operated since April, 2006. System availability and effectiveness of activity removal has been high. Operational issues of the system occurring during the first year of operation have been successfully solved.

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