Testing of an Iodine and Tritium Capture System for an NO_2-Based Tritium Pretreatment Process

Nuclear Technology Research and Development

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SUMMARY

The reprocessing of used nuclear fuel would release volatile radionuclides into the off-gas streams of a processing plant, including ^{3}H, ^{14}C, ^{85}Kr, and ^{129}I. One potential simplification to the management of the off-gas streams could be achieved through an efficient tritium pretreatment (TPT) step in which the UO_2 fuel is oxidized by either air or NO_2 before dissolution. The oxidation of the UO_2 fuel matrix will result in the release of tritium from the fuel. Upfront release of tritium from the fuel in a pretreatment step with subsequent tritium abatement can minimize or eliminate the distribution of tritium throughout a plant. This can decrease or eliminate the need for tritium capture on multiple off-gas streams and prevent distribution to the aqueous inventory. The use of NO_2 as the oxidant in an advanced tritium pretreatment (ATPT) allows for the oxidation to be performed at lower temperatures and may also result in the quantitative release of iodine from the fuel.

The ATPT off-gas will contain up to 75 vol% NO_2 gas. Spencer et al. (2017) identified the most promising iodine sorbent as silver nitrate-impregnated alumina (AgA) and the most promising tritium sorbents as 3Å molecular sieve (3Å MS) and silica gel for radionuclide capture from ATPT off-gas streams. An initial series of tests were conducted in fiscal years 2017–2018 (Jubin et al. 2018a) with the intent of demonstrating the recovery of iodine and tritium using these sorbents from a recirculating gas stream that simulated the NO_2 recycle loop envisioned for the ATPT system. These initial tests showed low tritium recoveries and significant variations in the iodine recovery on the AgA. Ultimately it was concluded, following disassembly of the system, that corrosion within the system was partially the cause of the poor recovery of both species. All components of that system were disposed of at the end of the analysis.

One of the conclusions drawn from the initial tests pointed to the reconsideration of the materials of construction. Based on that recommendation a new test bed for the advanced TPT off-gas system, that addresses the issues identified in the fiscal years 2017 and 2018 scoping tests, was designed and constructed. The test bed used high nickel alloys wherever possible to address the previously observed corrosion issues, a redesigned surge tank to permit ease of purging, an improved feed injection system and a more modular system design that allowed testing of incremental increases in system complexity to allow the isolation of any element that results in changes to recovery of target species.

Accompanying that design, a multifaceted test plan was completed (Jubin and Spencer 2018b). The overarching goals of this test series were two-fold. The first goal was to demonstrate the ability to recover both the iodine and tritium that would be released during TPT in a manner that the releases from the fuel can be quantified. The second goal was to demonstrate that the combined recovery system could operate in any of the three proposed TPT modes (once-through air or O_2, recirculating air or O_2, or recirculating NO_x).

A total of 22 tests were completed. Ten tests examined tritium behavior under varying operating parameters, 8 tests examined iodine behavior under varying operating parameters, and 4 tests examined both tritium and iodine behavior. Silica gel was used for the tritium sorbent, and AgA or Ag-mordenite (AgZ) was used as the iodine sorbent. The following conclusions were drawn:

- Full recovery is achieved for both sorbates when no sorbent is in place indicating no accumulation within the test system.
- In even the simplest of the configurations (i.e., no sorbents in the columns), iodine is more difficult to transport and recover from the system than tritium.
- Analysis of the AgA iodine sorbent beds from the integrated tritium and iodine tests indicate quantitative iodine recovery can be achieved (within anticipated experimental errors).

- This system may provide a means for the quantification of the iodine released by the ATPT process within the experimental uncertainties. This would support 0.33 to 1.00 kg demonstrations of the ATPT process with actual used nuclear fuel.
- Iodine penetrated further into AgZ than into AgA when operated in gas recirculation mode.
- Tritium adsorbs to and cannot be fully recovered from the 10-cm column of silica gel at regeneration temperatures up to 140 °C.
- Tritium adsorbs to and cannot be recovered from the 10-cm column of AgA at desorption temperatures up to 200 °C.
- Co-adsorbed tritium was recovered to a greater extent from AgZ than from AgA, but recovery was not complete.
- No visible corrosion was observed in the test system.

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ACRONYMS

3Å MS	3 angstrom molecular sieve
AgA	$AgNO_3$-impregnated alumina
AgZ	silver exchanged mordenite
ATPT	advanced tritium pretreatment
ATPTOG	advanced tritium pretreatment off-gas
DOG	dissolver off-gas
EG	ethylene glycol
HFIR	High Flux Isotope Reactor
IC	iodine capture [phase]
IP	iodine purge [phase]
MURR	University of Missouri Research Reactor
NAA	neutron activation analysis
ORNL	Oak Ridge National Laboratory
TC	tritium capture [phase]
TPTOG	tritium pretreatment off-gas
TPT	tritium pretreatment
TR	tritium sorbent regeneration or desorption [phase]

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TESTING OF AN IODINE AND TRITIUM CAPTURE SYSTEM FOR AN NO_2-BASED TRITIUM PRETREATMENT PROCESS

1. INTRODUCTION

The reprocessing of used nuclear fuel will release four volatile radionuclides into the off-gas streams of a processing plant. These radionuclides are ^{3}H, ^{14}C, ^{85}Kr, and ^{129}I. For a reprocessing plant sited within the United States and governed by US regulations, the abatement of some or all these radionuclides from the plant off-gas will be required before discharge of that gas to the environment. In some cases, the level of abatement required is significant (Jubin et al. 2012).

Traditional aqueous reprocessing dissolves sheared used fuel in hot nitric acid solution. As the fuel dissolves, the volatile radionuclides contained within the fuel are evolved into the dissolver off-gas stream. There is also some fraction of the volatile radionuclides that remain in the dissolver solution, and those radionuclides are transferred to subsequent processing steps and to the process off-gas streams generated by those processes. The distribution of the radionuclides into multiple liquid and gaseous streams results in the need to manage volatile radionuclide emissions from multiple off-gas streams associated with the reprocessing plant.

A potential simplification of the off-gas management system can be achieved through an efficient tritium pretreatment (TPT) step in which the UO_2 in the used fuel is oxidized before dissolution. Oxidation of the UO_2 fuel matrix results in the release of the tritium contained within the fuel matrix. Traditional TPT is conducted by oxidizing the fuel with a dry O_2-bearing gas stream at high temperatures. Upfront release of tritium from the fuel in a pretreatment step with subsequent tritium abatement can minimize or eliminate the distribution of tritium throughout a plant. This can decrease or eliminate the need for tritium capture on multiple off-gas streams and prevent distribution to the aqueous inventory.

An advanced variant of the O_2-based TPT process uses NO_2, a stronger oxidant, as the gaseous reagent. The use of NO_2 allows TPT to be performed at lower temperatures and may result in the quantitative release of iodine from the fuel. Upfront removal and abatement of iodine and tritium could result in a substantial simplification of the off-gas treatment processes. The advanced tritium pretreatment (ATPT) off-gas (ATPTOG) will contain up to 75 vol% NO_2 gas. Very little data is available on the performance of traditional iodine sorbents, such as silver-exchanged zeolites, and traditional tritium adsorbents, such as molecular sieves, under the highly oxidizing conditions present in the ATPTOG system.

A previous report (Spencer et al. 2017) identified the most promising iodine sorbent as silver nitrate–impregnated alumina (AgA) and the most promising tritium sorbents as 3Å molecular sieve (3Å MS) and silica gel for radionuclide capture from ATPTOG streams. The reaction between silver nitrate ($AgNO_3$) and I_2 to form AgI is thermodynamically favorable, and minerals impregnated with $AgNO_3$ have been used for I_2 removal from the dissolver off-gas streams at the La Hague fuel reprocessing plant in France. It was also believed that the nitrated form of silver ($AgNO_3$) would be more resistant to reactions with NO_2. However, the dissolver off-gas stream contains only ~2 vol% NO_x gaseous species, far below the levels expected in the ATPTOG stream.

The preliminary testing of AgA, 3Å MS, and silica gel for iodine and tritium removal from ATPTOG streams is documented in *Assembly and Testing of a Tritium and Iodine Removal System for Use with Advanced Tritium Pretreatment*, NTRD-MRWFD-2017-000157 (Jubin et al. 2017a). Tests were conducted in fiscal year 2017 and early fiscal year 2018 (Jubin et al. 2018a) with the intent of demonstrating the recovery of iodine and tritium from a recirculating gas stream that simulated the NO_2 recycle loop envisioned for the ATPT system. These initial tests showed low tritium recoveries and significant variations in the iodine recovery on the AgA. These results were unexpected, and several additional tests were performed in an attempt to understand the low recoveries. These tests failed to fully explain the low recovery performance and ultimately prompted a full system disassembly to assess whether there was hold-up of tritium and iodine within the system. The surge tank, which had been reused from a previous experimental system, showed signs of corrosion, and smears taken at various locations were positive for tritium at multiple locations. Several corroded fittings were removed from the surge tank, and both iodine and tritium were found in the corrosion products. No corrosion was observed at other visually inspected locations and no other inspected locations smeared positive for tritium.

One of the conclusions drawn from those tests pointed to the reconsideration of the materials of construction. Although the surge tank used in the initial tests was constructed of stainless steel, which is considered compatible with NO_2 gas and N_2O_4 liquid, other more corrosion-resistant materials should have been used. Iodine is known to corrode stainless steel in the presence of water. A second possibility is that NO_2 itself contributed to the corrosion of the system. The surge tank, which was the only visibly corroded component, was not heated during testing.

Based on that recommendation a new system was designed (Jubin and Spencer 2018b) and built that used high nickel alloys wherever possible, a redesigned surge tank, improved feed injection ports, and a more modular system design that supported testing of the system with incremental increases in complexity to allow the isolation of any element that results in changes to recovery of target species. A multifaceted test plan was also developed (Jubin and Spencer 2018b) for the test system that included a total of 15 test variants.

The overarching goals of the tests described in that report were two-fold. The first goal was to demonstrate the ability to recover both the iodine and tritium, that would be released during TPT in a manner that the releases from used fuel can be quantified. The second goal was to demonstrate the combined recovery system that could operate on any of the three proposed TPT modes (once-through air or O_2, recirculating air or O_2, or recirculating NO_x). The tests were, in many ways, similar in nature to the initial series of tests, but they incrementally added complexity to the system.

2. TEST SYSTEM AND TEST PLAN

2.1 Test Loop

The experimental methodology was reported previously in Jubin et al. (2017a) and the initial tests were reported in Jubin et al. (2018a). The flow diagram is shown in Figure 1, and a photo of the system before heating tapes and insulation were applied, is shown in Figure 2. The primary loop of this system was constructed using high nickel alloy (Hastelloy® C-276) components. The system is modular in design such that additional components can be valved-in. This allowed the tests to be performed in a sequence of increasing system complexity with the intent of ensuring that any changes in the amount of either tritium or iodine that is recovered is due to specific changes in operating modes and not due to losses within the system. Unlike the previous test loop, this loop was designed to operate in three possible TPT modes (once-through O_2, recirculating air or O_2, or recirculating NO_2). A major change is in the design of the surge tank, which was a 1-L Parr Hastelloy reaction vessel with a dip leg on the gas inlet port extending to near the bottom of the tank to minimize short circuiting within the tank. The tank also has provisions for heating to ensure that the NO_2 would not condense within the tank. The NO_2 feed lines were Teflon, and the discharge connection between the Hastelloy test system and the liquid scrubbers was also Teflon.

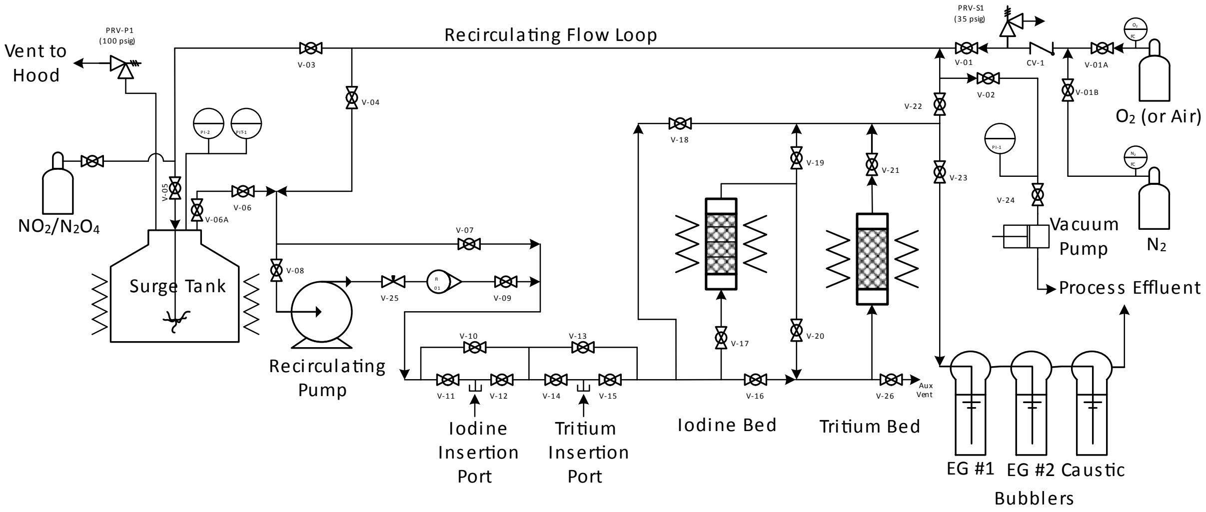

Figure 1: Schematic of iodine and tritium sorbent test system.

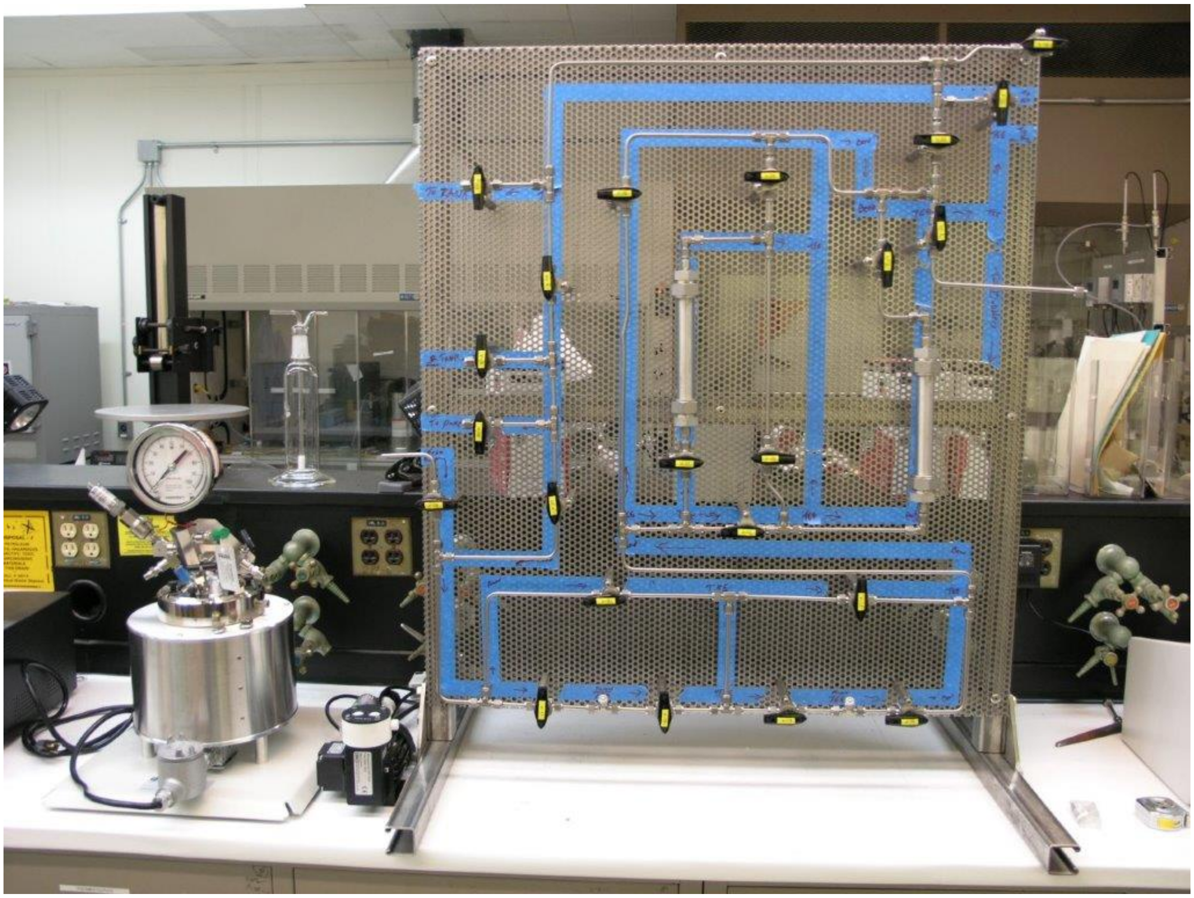

Figure 2: The iodine and tritium sorbent test system before installation in the radiological hood and placement of the heating tapes and insulation.

A total of seven individually controlled heating zones were provided as shown in Figure 3. Tritiated water and/or iodine was introduced to the system as required for a specific test through a modified loading system. This modified system is shown in Figure 4. This feed system was installed in the line between the recirculating pump and the iodine and tritium trapping systems. This feed system includes a valved gas bypass line and two sections, each containing a charging port. The charging sections had blocking valves at each end and a removable cap on a tee in the middle of the charging section. Before a test, with the bypass valves open and the blocking valves for the charging sections closed, the caps were removed, and a known amount of material was introduced into the loading chambers and the caps were replaced. As with the rest of the system lines, the charging chambers were heat traced, but they were on a separate control that allowed them to be heated independently. This permits the charging system to be loaded cold while the balance of the system is heating to the desired operating temperature. Once the charging system is loaded and recapped, it can be heated at the appropriate time. At the desired start time for iodine and/or tritium introduction into the flowing gas stream, the blocking valves on the individual charging sections are opened, and the associated bypass valve is closed to allow the gas stream to pass through the charging section. The design permits direct flow of the full stream through either or both of the charging sections.

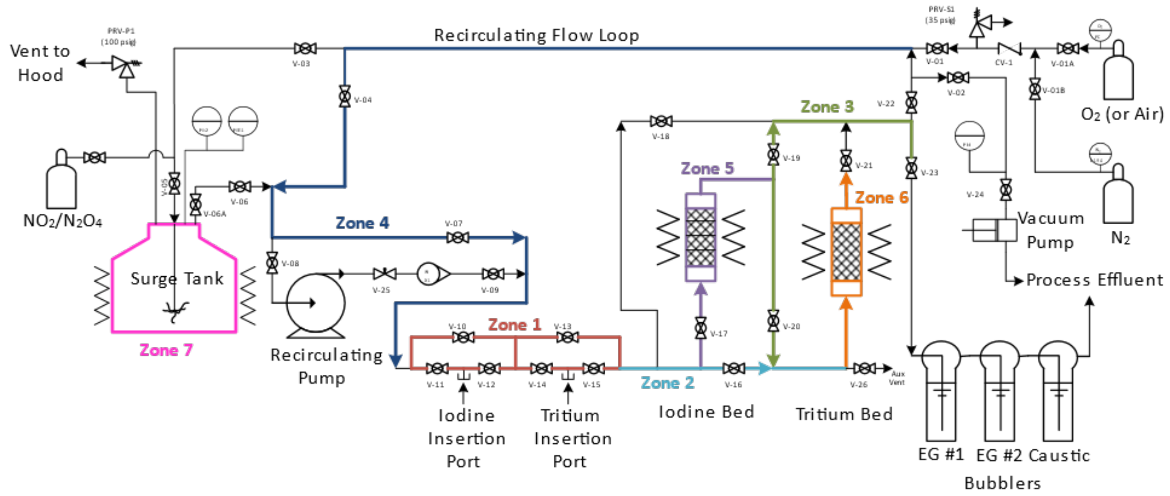

Figure 3: Heating zones for the test system.

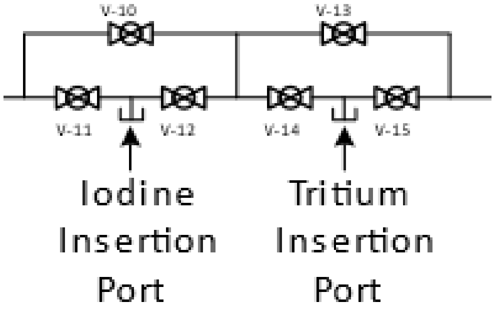

Figure 4: Sample charging system design.

2.2 Test Plan

A total of 15 test conditions were planned (Jubin and Spencer 2018b), and the details of each of the planned tests are summarized in Table 1. The test identification numbers used in this experimental effort reflect the intent of the planned individual experiments laid out in the report by Jubin and Spencer (2018b). However, based on knowledge gained during this experimental program, some of the experimental details were modified, and selected experiments were omitted.

Seven of the planned tests were to be conducted using deuterium as a stable surrogate for tritium. While the use of deuterium tended to simplify the radiological aspects of the work, the additional time and cost of the subsequent deuterium analysis ultimately outweighed the benefits provided, and a decision was made to use tritium in all cases. As a result, tests ATPT19-12 and ATPT19-14 were omitted as they were now redundant with the actual tritium counterparts.

Additionally, after the completion of test ATPT19-5 and the receipt of analytical data that indicated the silica gel appeared to retain a significant amount of the injected tritium, silica gel was omitted from subsequent tests. Because the data indicated that silica gel was effective in the trapping mode, the tritium could just as easily be trapped on the silica gel if it could be demonstrated that the tritium either passed through the iodine sorbent bed of AgA or could be subsequently desorbed from the AgA and transferred to the effluent bubblers.

Tests ATPT19-8 through -10 were omitted because the objectives of these tests were accomplished in tests ATAP-19-11 through -15. Two other changes from the planned tests were the elimination of the test using 3Å MS for tritium capture (ATPT19-15b) due to the limited tritium desorption from the AgA and replacing the repeated pressurization depressurization of the surge tank (ATPT19-4b) in favor of the use of a flowing sweep gas to purge the system. ATPT19-15b was replaced with a repeat of ATAP19-15a using AgZ rather than AgA as the sorbent.

2.3 Test Phases and System Configurations

The tests were designed to include the following phases:

1. Iodine capture (or simulated iodine capture) (IC)
2. Tritium capture (TC)
3. Tritium sorbent regeneration (TR)
4. Iodine sorbent purge (IP)

Between each of the phases the valve positions and operating parameters were changed to provide the appropriate flow paths for the gas stream. Several of the tests (e.g., once-through) did not include one or more of the phases, and some tests combined the tritium regeneration and iodine purge into one operation. Brief descriptions of the phases are presented below. In these descriptions and associated figures, the heavy black lines are the primary flow paths. Depending on the test system configuration, portions of the system may be bypassed. Alternate pathways are represented by the red (once-through) and blue (recirculation) lines in Figures 5 and 6.

Table 1: Descriptions of the planned tests.

Test ID	Sorbent (iodine / tritium)	Mode of operation	Carrier gas	Sorbates	Tritium desorption	Notes
ATPT19–1	None / None	Once-through	Air	Deuterated Water	N/A	Establish deuterium recovery in ethylene glycol baseline
ATPT19–2	None / Silica gel	Once-through	Air	Deuterated Water	90 °C	Establish deuterium desorption recovery in ethylene glycol baseline
ATPT19–3	None / Silica gel	Recirculation	Air	Deuterated Water	90 °C	Establish deuterium desorption recovery in ethylene glycol baseline
ATPT19–4a	None / Silica gel	Recirculation / with Surge tank	Air	Deuterated Water	90 °C	Establish deuterium desorption recovery in ethylene glycol baseline Compare deuterium purge / capture methods
ATPT19–4b	None / Silica gel	Recirculation / with Surge tank	Air	Deuterated Water	90 °C	Establish deuterium desorption recovery in ethylene glycol baseline Compare deuterium purge / capture methods
ATPT19–5	None / Silica gel	Recirculation / with Surge tank	NO_2	Deuterated Water	90 °C	Establish deuterium desorption recovery in ethylene glycol baseline
ATPT19–6	None / None	Once-through	Air	Iodine	N/A	Establish iodine recovery in ethylene glycol and caustic baseline
ATPT19–7	AgA / None	Once-through	Air	Iodine	N/A	Establish iodine recovery on AgA in ethylene glycol and caustic baseline
ATPT19–8	AgA / None	Recirculation	Air	Iodine	N/A	Establish iodine recovery on AgA
ATPT19–9	AgA / None	Recirculation / with Surge tank	Air	Iodine	N/A	Establish iodine recovery on AgA
ATPT19–10	AgA / None	Recirculation / with Surge tank	NO_2	Iodine	N/A	Establish iodine recovery on AgA
ATPT19–11	AgA / Silica Gel	Once-through / with Surge tank	Air	Iodine/ Deuterated Water	90 °C	Establish iodine recovery on AgA deuterium desorption recovery in ethylene glycol baseline
ATPT19–12	AgA / Silica Gel	Recirculation / with Surge tank	Air	Iodine/ Deuterated Water	90 °C	Establish iodine recovery on AgA deuterium desorption recovery in ethylene glycol baseline
ATPT19–13	AgA / Silica Gel	Recirculation / with Surge tank	Air	Iodine/ Tritiated Water	90 °C	Establish iodine recovery on AgA deuterium desorption recovery in ethylene glycol baseline
ATPT19–14	AgA / Silica Gel	Recirculation / with Surge tank	NO_2	Iodine/ Deuterated Water	90 °C	Establish iodine recovery on AgA tritium desorption recovery in ethylene glycol baseline
ATPT19–15a	AgA / Silica Gel	Recirculation / with Surge tank	NO_2	Iodine/ Tritiated Water	90 °C	Establish iodine recovery on AgA tritium desorption recovery in ethylene glycol baseline
ATPT19–15b	AgA / 3Å MS	Recirculation / with Surge tank	NO_2	Iodine/ Tritiated Water	250 °C	Establish iodine recovery on AgA tritium desorption recovery in ethylene glycol baseline

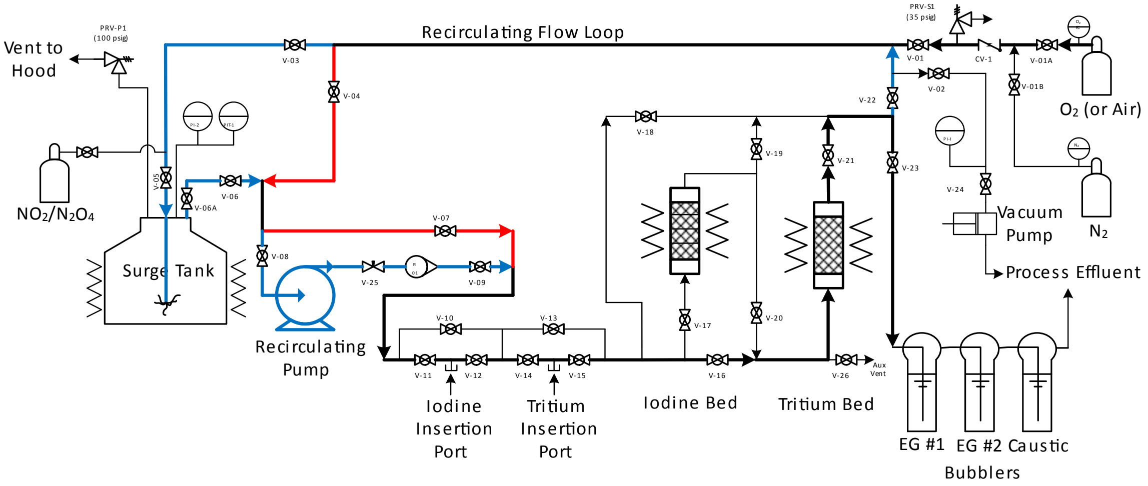

Figure 5: Gas flow path (bolded) during tritium capture and regeneration phase. Tritium was ultimately captured by both AgA in the iodine bed and silica gel in the tritium bed.

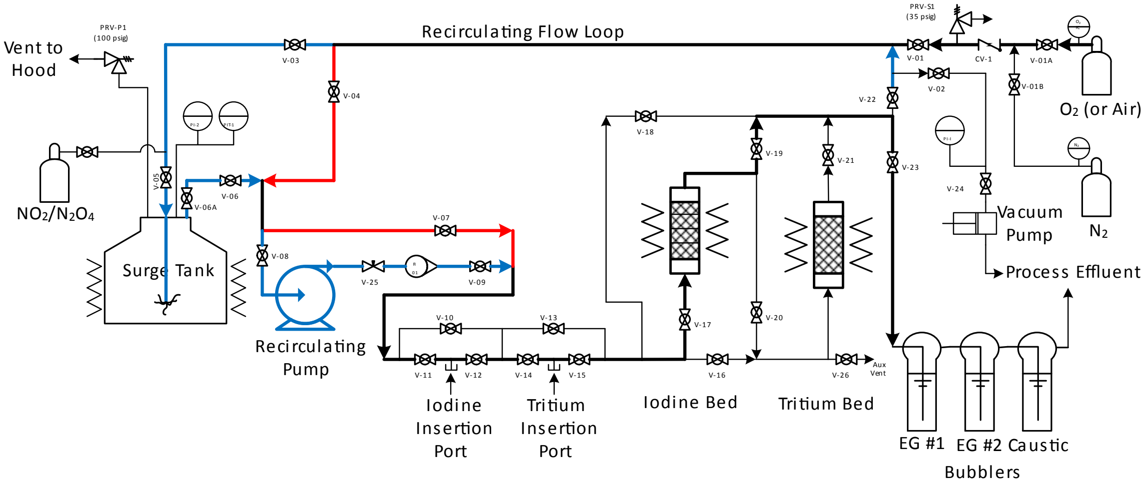

Figure 6: Gas flow path (bolded) during iodine sorbent loading and purge phase.

2.3.1 Iodine Capture Phase

This phase typically involves the initial loading of the iodine trap. This phase is simulated or bypassed in tests 1–5 when there is no AgA sorbent in place.

2.3.2 Tritium Capture Phase

After IC and recirculation, the recirculation pump is turned off, and the tritium bed is heated to 30–40 °C. The flow path is altered so that the gas stream does not pass through the iodine bed but passes through the tritium bed and then out of the system through bubblers of ethylene glycol (EG) and/or sodium hydroxide (NaOH) as shown in Figure 5.

2.3.3 Tritium Sorbent Regeneration Phase

The temperature of the tritium bed is raised to the selected regeneration temperature, and the bed is purged with oxygen at 0.5 L/min for at least 2 hours. The effluent from the tritium column flows through ethylene glycol scrubbers and/or NaOH scrubbers, with the scrubber solutions sampled between the TC and TR phases of testing. The flow path for the oxygen purge is shown in Figure 5.

2.3.4 Iodine Sorbent Purge

If needed, the system valving alignment is modified so that the tritium column is isolated from the rest of the system, and the iodine column is included in the flow path (Figure 6). The iodine column is held at 200–250 °C and purged with oxygen at 0.5 L/min for at least 2 hours. The effluent passes through ethylene glycol and/or NaOH scrubbers. The scrubber solutions are sampled between the TR and IP phases of testing. During tests with AgA as a sole sorbent, the tritium regeneration (TR) and iodine purge (IP) phases were accomplished simultaneously.

3. MATERIALS AND METHODS

3.1 Iodine and Tritium Sorbents

The AgA used in the iodine capture column was prepared at Oak Ridge National Laboratory (ORNL) by combining 500 g of 1.18–1.40 mm diameter activated alumina desiccant spheres (Sorbent Technologies) with 380 mL of 1.936 *M* $AgNO_3$ solution. This mixture was heated to a maximum temperature of 125 °C for 9 hours under vacuum. After this process was completed, the dried alumina spheres weighed 653.3 g, indicating deposition of $AgNO_3$ in the porous structure of the spheres.

Silver mordenite was obtained from Molecular Products in an engineered pelletized form (Ionex-Type Ag 900 E16). It contains 11.9% silver by weight and has a 0.16 cm pellet diameter. Before use in testing, the sorbent material was reduced by extended exposure to a hydrogen–nitrogen blend gas at elevated temperature. Details of this procedure can be found in Anderson et al. (2012).

Before the start of each test, the iodine and/or silica gel column were loaded with fresh sorbent for all tests except for tests ATPT19-1 and -6, which required empty columns. The iodine column was separated into three to four discrete segments with layers of quartz wool and stainless-steel mesh disk between the segments. The total length of the active (AgA) iodine bed was ~10 cm. The tritium column contained silica gel of approximately 6–12 mesh size, with a bed depth of 10 cm.

The planned loading temperature for the silica gel was 40 °C, while it was 150 °C for the AgA. The silica gel was intended to be used as a regenerative sorbent, in which the adsorbed tritium (as tritiated water or tritiated nitric acid) is loaded onto the sorbent at operating temperature and then subsequently desorbed at high temperature into a small-volume gas stream for recovery and conversion to a waste form. The manufacturer-recommended desorption temperature is >90 °C.

3.2 Tritium Feed

Each test involving tritium used 100 µL of tritiated water. This amount of water is significantly greater than the amount expected of HTO in 1 kg of actual used nuclear fuel with a burnup of 30 GWd/MTIHM and 30-year cooling time (~0.058 µL HTO or ~1.3 µL of combined $H_2O + HDO + HTO$ from the fuel) (Jubin and Strachan 2015), but this amount reflects the need to add 100 fold more water to achieve a tritium decontamination factor of 100 at these very low concentrations.

The tritium charging section is an isolated section of line with a removable fitting to allow for the introduction of the sample. Before the start of the test, with the charging section of the system unheated, the valves were configured to bypass both charging sections. The cap on the tritium charging section was opened and a known amount tritiated water pipetted into the port, and the cap was replaced. The charging section of the system was then heated to the desired temperature. At the desired time in a specific run, the two block valves at the ends of this charging section were opened, and the bypass valve for that section was closed. With the load chamber heated, the tritiated water was expected transfer into the gas recirculation loop quickly and completely.

3.3 Iodine Feed

Iodine was introduced as crystalline I_2. Approximately 0.13 g was used in each test. This amount of iodine is based on the expected quantity of total iodine in 0.7 kg of used nuclear fuel with a burnup of 30 GWd/MTIHM and 30-year cooling time (Jubin and Strachan 2015). A charging system similar to the one used for tritium was employed. With the load chamber heated, the iodine was also expected transfer into the gas recirculation loop quickly and completely.

3.4 Analytical Methods

Tritium activity in the scrubber solutions and on the sorbent were measured at ORNL by a scintillation counter with a limit of detection of 0.52 Bq/mL. Iodine concentrations in the scrubber solutions were measured by ICP-MS at ORNL with a limit of detection of 3.00E-01 µg/mL. Iodine concentrations on the sorbent were measured using neutron activation analysis (NAA) at the University of Missouri Research Reactor (MURR). Two tritium standard solutions were prepared by adding 100 µL of tritiated water (0.486 mCi/mL) to 200 mL ethylene glycol. The calculated activity of these standards is 8.99E+03 Bq/mL based on the reported activity in the stock solution. The analytical laboratory analysis of the standard solutions was 9.86E+03 ± 1.50E+02 Bq/mL (average of six analyses). This later value was used as the basis for tritium recovery. Three iodine standards were prepared by adding ~0.13 g of iodine crystals to 200 mL of caustic or EG. The two caustic iodine standard concentrations were calculated to be 6.63E+02 µg/mL (caustic 1) and 6.75E+02 µg/mL (caustic 2), and the analytical lab returned values of 6.79E+02 ± 1.51E+01 µg/mL (102% recovery) and 7.11E+02 ± 2.84E+01 µg/mL (105% recovery). The EG iodine standard concentrations were calculated to be 6.52E+02 µg/mL and the analytical lab returned values of 7.16E+02 ± 7.16 E+01 µg/mL (110% recovery) and 5.92E+02 ± 5.92E+01 µg/mL (90.8% recovery).

4. TEST DESCRIPTIONS

A total of 15 categories of tests were planned. Tests ATPT19-1–5 were all performed with tritiated water instead of the planned deuterated water to facilitate more rapid turnaround on analytical results. Tests ATPT19-6–10 were planned to be iodine only, but tests 8–10 were accomplished as part of the ATPT19-11–15 series. This change was due to the longer turnaround times associated with the NAA of iodine that were performed at the MURR. MURR was used because of the prolonged outage at the ORNL High Flux Isotope Reactor. A summary of the tests conducted is presented in Table 2.

These test plans were based on the assumption that the selected sorbent would perform as expected and that the limited recovery in the initial tests was the result of the corrosion noted within the system. If recovery issues were observed, the modular design would allow additional tests to isolate the origin of the losses and/or to replace the iodine and/or tritium capture beds with an alternate system.

AgA was tested as a potential iodine and tritium sorbent at a loading temperature of 150–200 °C and silica gel was tested as potential tritium sorbent with a loading temperature of 30–40 °C. While the initial test plan listed only silica gel as a tritium sorbent, preliminary tests showed that, when heated to 200 °C, the AgA loses up to ~4 wt% moisture, and therefore could adsorb significant tritium after it is dehydrated. The iodine purge (IP) from the AgA was tested at 200–250 °C and regeneration of tritium (TR) from the silica gel was tested at 90–140 °C.

Table 2: Summary of test conditions.

Test ID	**Seq. no.**	**Sorbent (iodine/ tritium)**	**Mode of operation**	**Carrier gas**	**Sorbate**	**Sorption temp.**	**Sorption flow rate (L/min)**	**Desorption temp.**	**Desorption flow rate (L/min)**	**Recovery solution**	**Notes**
ATPT19-1a	1	None/None	Once-through	O_2	tritium	40 °C	0.25	N/A	N/A	EG + NaOH	Establish tritium baseline over 30 min.
ATPT19-1b	2	None/None	Once-through	O_2	tritium	40 °C	0.25	N/A	N/A	EG + NaOH	Establish tritium baseline over 30 min.
ATPT19-1c	3	None/None	Once-through	O_2	tritium	40 °C	0.25	N/A	N/A	EG + NaOH	Establish tritium baseline over 30 min.
ATPT19-1d	7	None/None	Once-through	O_2	tritium	75 °C	0.25	N/A	N/A	EG	Establish tritium baseline over 2 hours
ATPT19-2a	4	None/Silica gel	Once-through	O_2	tritium	40 °C	0.25	90 °C	0.5	EG + NaOH	Establish tritium recovery from silica gel
ATPT19-3a	5	None/Silica gel	Recirculation 30 min	O_2	tritium	40 °C	0.25	90 °C	0.25	EG + NaOH	Establish tritium recovery from silica gel
ATPT19-3b	6	None/Silica gel	Desorption #2	O_2	tritium	N/A	N/A	140 °C	0.25	EG + NaOH	Desorbed tritium from 3a in reverse flow
ATPT19-3c	10	AgA/None	Recirculation 30 min	O_2	tritium	150 °C	0.25	200 °C	0.25	EG + NaOH	Establish tritium recovery from AgA
ATPT19-4a	8	None/Silica gel	Recirculation w/ Surge Tank	O_2	tritium	30 °C	0.25	140 °C	0.25	EG	Establish tritium recovery (desorbed in reverse flow)
ATPT19-5a	9	None/Silica gel	Recirculation w/ Surge Tank	NO_2	tritium	30 °C	0.25	140 °C	0.25	EG + NaOH	Establish tritium recovery (desorbed in reverse flow)
ATPT19-6a	11	None/None	Once-through	O_2	iodine	150 °C	0.25	N/A	N/A	NaOH	Establish iodine baseline over 30 min.
ATPT19-6b	12	None/None	Once-through	O_2	iodine	150 °C	0.25	N/A	N/A	NaOH	Establish iodine baseline over 30 min.
ATPT19-6c	13	None/None	Once-through	O_2	iodine	150 °C	0.25	N/A	N/A	EG + NaOH	Establish iodine baseline over 30 min.
ATPT19-6d	15	None/None	Once-through	O_2	iodine	150 °C	0.25	N/A	N/A	EG + NaOH	Establish iodine baseline over 4 hours
ATPT19-6e	17	None/None	Once-through	O_2	iodine	200 °C	0.5	N/A	N/A	EG + NaOH	Establish iodine baseline over 3 hours
ATPT19-6f	20	None/None	Once-through	O_2	iodine	N/A	0.5	N/A	N/A	NaOH	Establish iodine baseline without column
ATPT19-6g	22	None/None	Once-through	O_2	iodine	150 °C	0.25	N/A	N/A	NaOH	Establish iodine baseline over 5 hours

Test ID	**Seq. no.**	**Sorbent (iodine/ tritium)**	**Mode of operation**	**Carrier gas**	**Sorbate**	**Sorption temp.**	**Sorption flow rate (L/min)**	**Desorption temp.**	**Desorption flow rate (L/min)**	**Recovery solution**	**Notes**
ATPT19-7a	14	AgA/None	Once-through	O_2	iodine	150 °C	0.25	N/A	N/A	NaOH	Establish iodine sorption onto AgA
ATPT19-11a	16	AgA/None	Once-through	O_2	iodine + tritium	150 °C	0.25	250 °C	0.5	EG + NaOH	Establish iodine and tritium sorption onto AgA
ATPT19-13a	18	AgA/None	Recirculation w/ Surge Tank	O_2	iodine + tritium	150 °C	0.25	200 °C	0.5	EG + NaOH	Establish iodine and tritium sorption onto AgA
ATPT19-15a	19	AgA/None	Recirculation w/ Surge Tank	NO_2	iodine + tritium	150 °C	0.25	200 °C	0.5	NaOH	Establish iodine and tritium sorption onto AgA
ATPT19-15b	21	AgZ/None	Recirculation w/ Surge Tank	NO_2	iodine + tritium	150 °C	0.25	200 °C	0.5	NaOH	Establish iodine and tritium sorption onto AgZ

Note: Seq. no. refers to the order the tests were completed.

The AgA was loaded into the column in discrete sections, each separated by glass wool and stainless-steel mesh. For most runs, the lowermost section was 1–2 cm in height, the middle section was 2–3 cm in height, and the top section was ~6 cm in height. A weighed amount of sorbent was used to produce a 10 cm tall bed in the column. Tests often used more than one scrubber in series with combinations of scrubber types (e.g., 2 EG, 2 caustic, EG followed by caustic, etc.). Following the completion of the proposed test matrix, a final test, (15b), was run using Ag-mordenite (AgZ) as a sorbent in place of AgA.

The once-through tests involved loading the sorbate and flowing it through the sorbent bed with O_2 as a carrier gas before discharging to one or more scrubbers. The recirculation mode operated with O_2 or NO_2 / O_2 mixture as a carrier gas, using an in-line pump to circulate the carrier gas and sorbate throughout the system for 30 minutes with or without the surge tank in the flow circuit. After recirculation, the carrier gas was discharged to one or more scrubbers.

4.1 Descriptions of the Individual Tests

4.1.1 Test ATPT19-1: Once-through Tritium Recovery

The objective of test ATPT19-1 was to demonstrate the quantitative or baseline recovery of the injected tritium by the ethylene glycol (EG) traps. The flowsheet for this test is shown in Figure 7, with thickened lines indicating the flow path. Tritiated water was introduced in the system through the tritium port described above. Both the tritium trap and the iodine trap sections were in place but did not contain sorbent. The O_2 carrier gas was introduced at a rate of 0.25 L/min and passed through the system, bypassing the surge tank and recirculation pump and passing through the charging section and empty tritium bed before passing through the glycol traps and caustic trap.

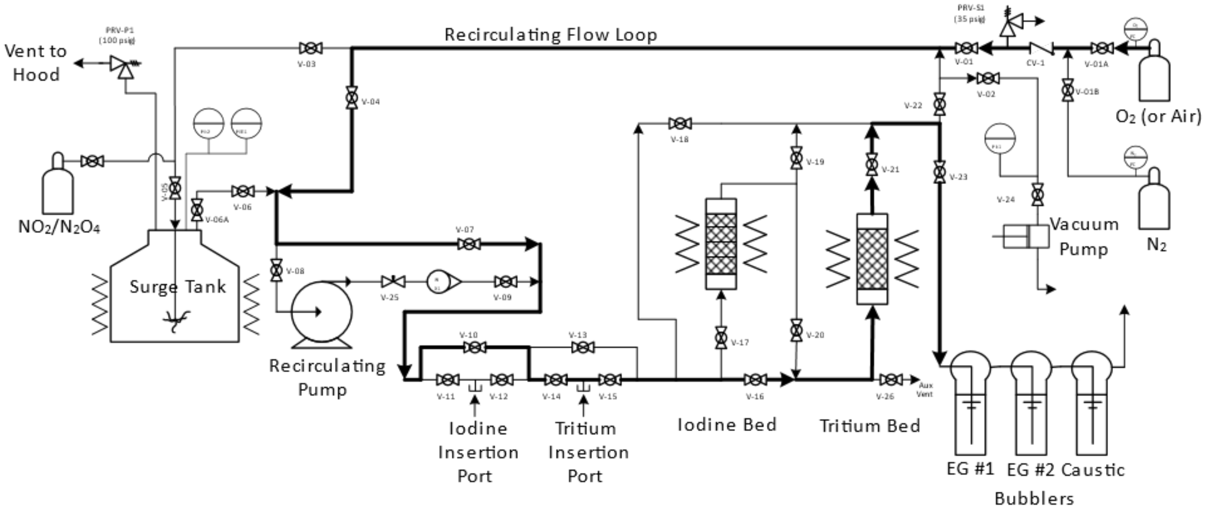

Figure 7: Flow diagram for tests ATPT19-1 and -2. Thickened lines indicate the flow path.

This test was repeated three times (ATPT19-1a, ATPT19-1b, and ATPT19-1c) in 1 day at 40 °C for 30 minutes of total flow per test. The tritium recovery in each of the scrubbers is shown in Figure 8. The results from the first three tests indicate that 30 minutes is not adequate time to capture all tritium introduced into the system. Test ATPT19-1a showed only ~50% recovery, and tests -1b and -1c showed closer to 87% recovery. Because tests ATPT19-1b and -1c could have recovered residual tritium from test 1a, test ATPT19-1 was repeated a fourth time (ATPT19-1d), at a later date, after the system had been purged for 3 hours. Test ATPT19-1d ran at 75 °C for 2 hours into only one EG scrubber. Test 1d confirmed that only ~50% of the tritium is recovered in 30 minutes, and 84% of the tritium can be recovered after 2 hours. The tritium recovered in the second EG scrubber and caustic scrubber showed that the initial EG scrubber was efficient enough to capture all the tritium and as a result the number of scrubbers used in tests following the receipt of this data was reduced.

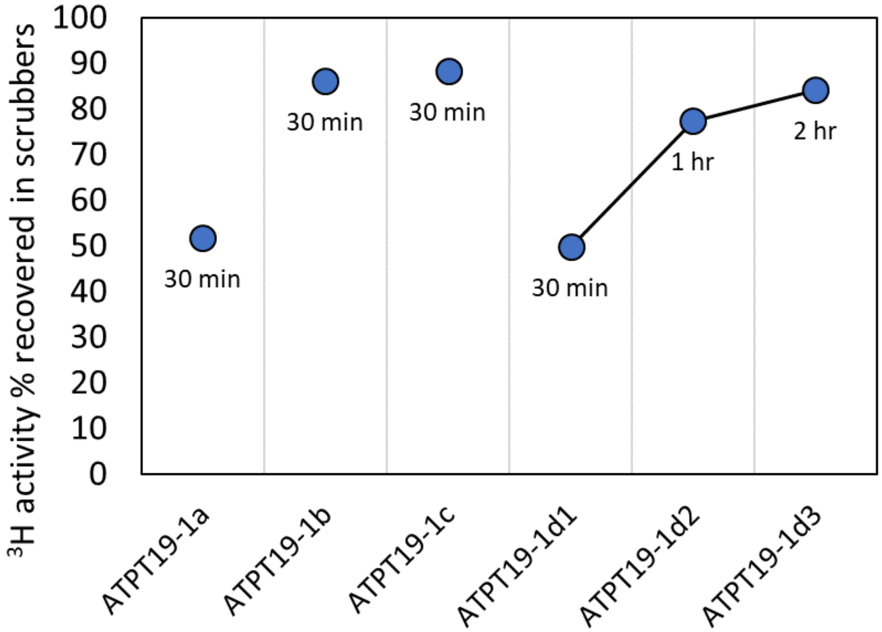

Figure 8: Percent of injected tritium recovered in scrubber(s) during tests ATPT19-1a–1d (once-through with no sorbent). Data points for tests 1a–1c represent the cumulative tritium from three scrubbers sampled at the end of each test.

4.1.2 Test ATPT19-2: Once-through Tritium Sorption/Desorption onto Silica Gel

The objective of test ATPT19-2 was to demonstrate the quantitative recovery of the injected tritium that is sorbed on the silica gel and subsequently recovered in the ethylene glycol bubblers. The flowsheet for this test is represented by Figure 7. Tritiated water was introduced in the system as was done in test ATPT19-1. The O_2 carrier gas was introduced at a rate of 0.25 L/min and passed through the system, bypassing the surge tank and recirculation pump, before passing through the charging section and tritium bed and exiting through the glycol bubblers. The system was operated in the loading mode for 30 minutes with the silica gel bed at 40 °C. At the end of the 30-minute loading period, the glycol traps were sampled to determine the amount of tritium that penetrated the bed. The tritium trap was regenerated using the same methods as used in the initial tests reported by Jubin et al. (2018a).

In the regeneration portions of those tests, the system was then sealed, and the temperature of the tritium bed was raised to the selected regeneration temperature (90 °C). After reaching the set point, the system was opened, and the bed was purged with oxygen at 0.5 L/min for 2 hours. The effluent from the tritium column passed through two ethylene glycol scrubbers and one NaOH scrubber, and the scrubber solutions were sampled between the collection and regeneration phases of testing.

The analytical results show that approximately 18% of the tritium passed through the silica gel during the sorption period, but almost no tritium was removed from the silica gel during the desorption phase.

4.1.3 Test ATPT19-3: Recirculation Tritium Sorption/Regeneration onto Silica Gel or AgA

The objective of test ATPT19-3 was to quantify the recovery of the injected tritium recirculated through the systems and then adsorbed on the silica gel. The silica gel was then regenerated to recover tritium in the ethylene glycol bubblers. The flowsheets showing the three main phases of this test are shown in Figures 9–11. Three variants of this test were performed, and the individual tests are described below. Figure 12 shows the results.

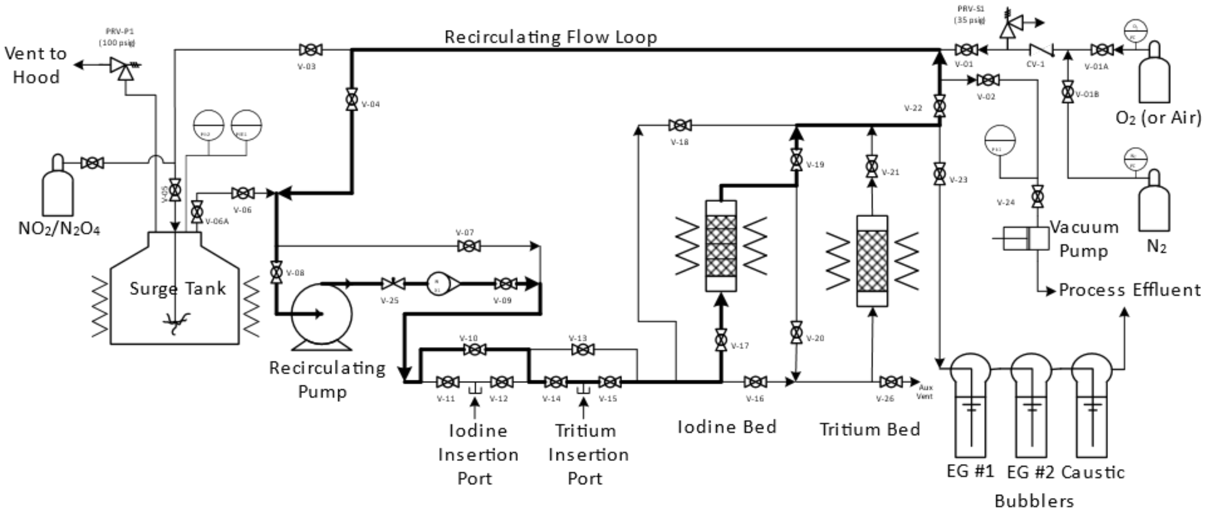

Figure 9: Flow diagram for Test ATPT19-3 part 1 (recirculation).

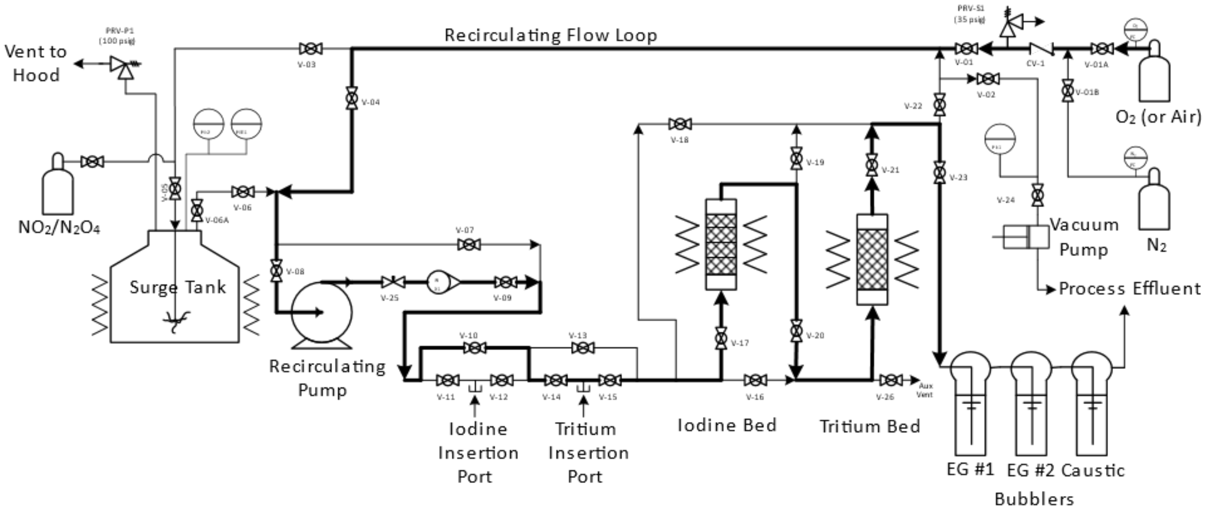

Figure 10: Flow diagram for Test APTP19-3 part 2 (load onto silica gel then flow to scrubbers).

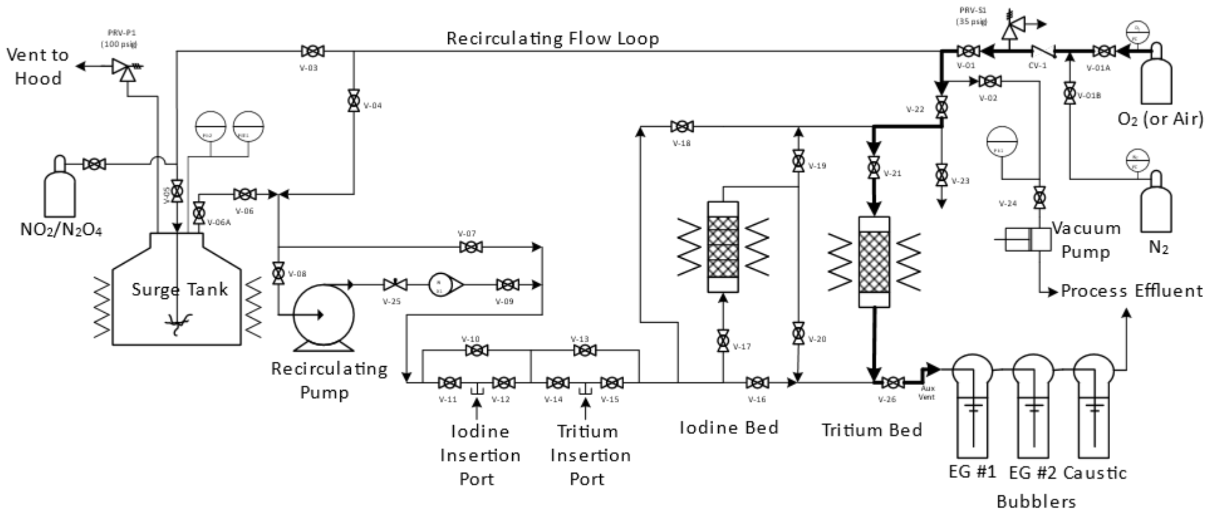

Figure 11: Flow diagram for Test ATPT19-3b and -5a (reverse flow through silica gel to recover tritium).

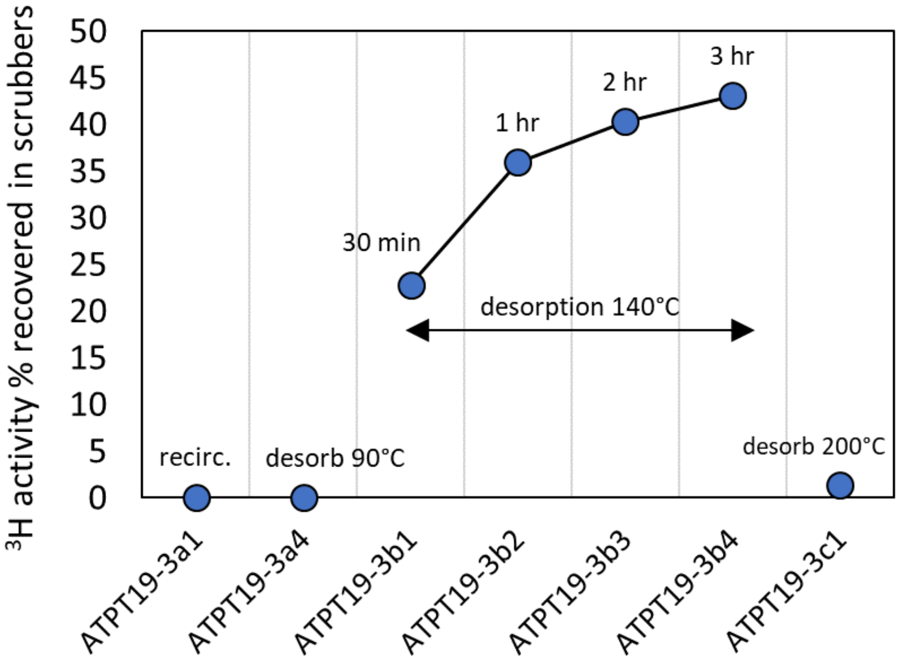

Figure 12: Percent of injected tritium recovered in scrubbers from test ATPT19-3a–3c (once-through tritium recovery from silica gel for tests 3a and 3b and from AgA for test 3c). Data points represent the cumulative tritium from each scrubber at that sampling stage. Test 3a was desorbed in the same flow direction as was used in the initial loading, and tests 3b and 3c were desorbed in the reverse flow configuration.

In test ATPT19-3a, silica gel was loaded into the column and dried with O_2 at 90 °C for 2 hours to prepare the system for operation. Tritium was then recirculated through the empty iodine column system in a simulated IC phase, bypassing the silica gel column, for 30 minutes. In test ATPT19-3a, the tritium-containing gas stream was then redirected to adsorb onto the silica gel bed for 20 minutes (operated at 40 °C), the TC phase. Any tritium that penetrated the silica gel was captured in three scrubbers (samples 3a1–3a3). In this test, no tritium was observed to have penetrated the silica gel. After sorption, the silica gel was desorbed, the TR phase, by heating to 90 °C and a flowing stream of O_2 at 0.25 L/min through the column for 2 hours. The effluent from the tritium column was routed to the scrubbers (samples 3a4–3a6). No tritium was observed in the samples taken from the scrubbers after desorption.

The test system was then reconfigured for test ATPT19-3b to flow O_2 backward through the silica gel column, under the assumption that tritium from test ATPT19-3a was loaded primarily on the inlet or lower portion of the silica gel bed. A second regeneration of the bed was conducted by heating the bed to 140 °C and flowing a stream of O_2 through the bed at 0.25 L/min for 3 hours This reverse flow resulted in the release of ~43% of the tritium that was introduced to the system in test ATPT19-3a (samples ATPT19-3b1–3b4).

Test ATPT19-3c examined the expected hold-up of tritium on the AgA and the subsequent recovery of tritium. Tritiated water was recirculated through the packed AgA column at 150 °C and 0.25 L/min for 30 minutes. After the 30-minute recirculation period, the flow was diverted to the EG scrubber, and the mode of operation was configured to once-through. The AgA column was then heated to 200 °C and purged with O_2 at a flow rate of 0.25 L/min for 80 minutes to desorb the tritium. The effluent was routed to an EG scrubber. The analytical results show extremely low tritium activity in the scrubber solution (sample 3c1), indicating near complete retention of tritium within the system, and the tritium was thought to be bound to the AgA. When the AgA was examined after completion of test 3c, significant fines were found to have formed, and most of the AgA was black and looked charred. This did not happen in future tests with the AgA (despite running at similar temperature conditions), and the cause of its disintegration during test 3c is unknown. This test was completed before it was discovered that the AgA releases up to 4 wt% moisture during heating.

4.1.4 Tests ATPT19-4: Recirculation and Surge Tank Operation with Tritium Sorption/Regeneration onto Silica gel Using O_2

The objective of test ATPT19-4 was identical to that of test ATPT19-3, but with the addition of the surge tank into the flow loop. The flowsheet for this test is shown in Figures 13 and 14. After loading into the column, the silica gel was dried at 120 °C. Oxygen and tritium were recirculated at a rate of ~0.25 L/min through the empty iodine bed, and the surge tank and recirculation pump ran in the iodine capture mode for 30 minutes. At the end of the 30-minute loading period, the iodine trap was isolated, and the carrier gas was redirected through the 30 °C silica gel column for 30 minutes in tritium capture mode. The tritium was then recovered from the silica gel under the reverse flow configuration at 140 °C for ~2.5 hours. (Note: Based on tritium recovery data from the initial tests, the adsorption and desorption temperatures and flow paths were altered from those used in tests ATPT19-3a and -3b to improve the tritium recovery from the system.)

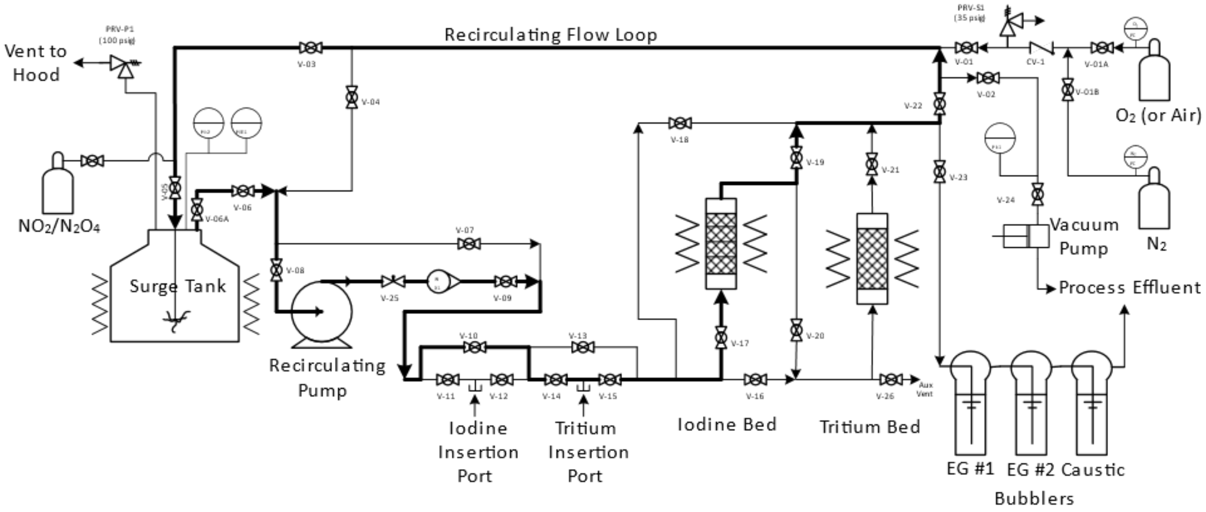

Figure 13: Flow diagram for Tests ATPT19-4 and -5 part 1 (recirculation through empty AgA column).

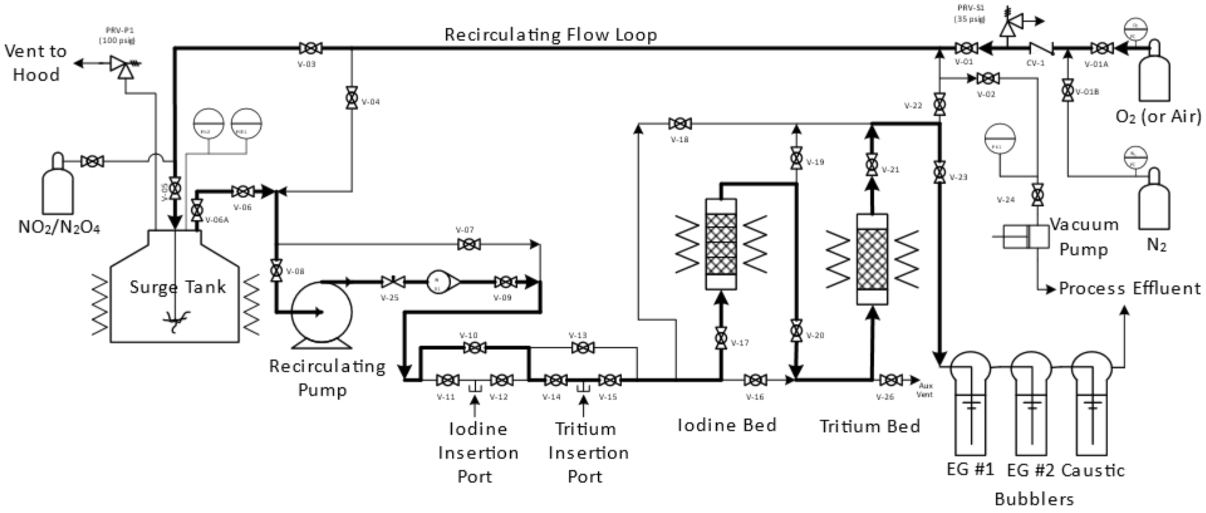

Figure 14: Flow diagram for Tests ATPT19-4 and -5 part 2 (tritium adsorption onto silica gel phase and gas sampling with bubblers).

Samples were collected in EG after the sorption phase (4a1) and after the desorption phase (4a2, 4a3), and the results are presented in Figure 15 along with the tritium recovery data from test ATPT19-5 that will be discussed in the next section. As can be seen, tritium did not penetrate the silica gel bed during the sorption phase. However, tritium was also not fully recovered from the test bed during the regeneration phase. Test 4 showed ~4% tritium recovery even with 2.5 hours in silica gel bed regeneration mode.

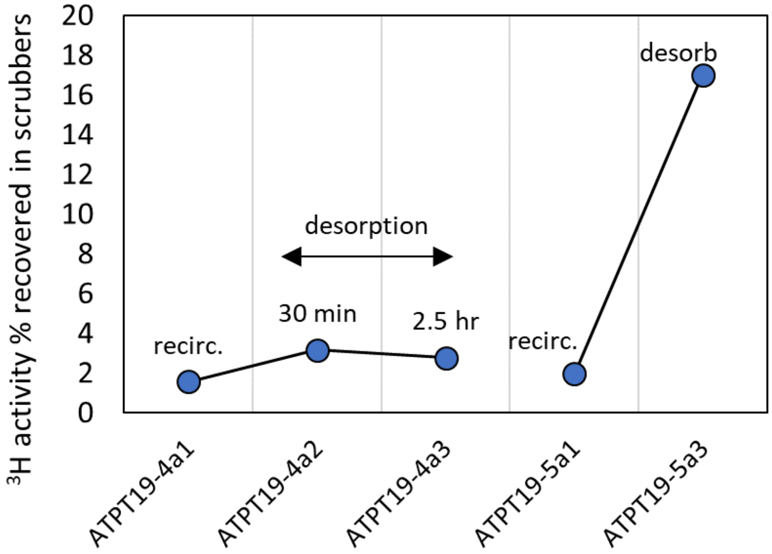

Figure 15: Percent of injected tritium recovered in the scrubbers during test ATPT19-4 (O_2 carrier gas) and -5 (NO_2 carrier gas). Data points represent the cumulative tritium from both scrubbers at the indicated sampling phase.

4.1.5 Tests ATPT19-5: Recirculation and Surge Tank Operation with Tritium Sorption/Regeneration of Silica gel using NO_2

The objective of test ATPT19-5 is the same as tests ATPT19-3 and ATPT19-4 but using ~45%–50% NO_2 instead of pure O_2 as the carrier gas. The flowsheet for this test is shown in Figures 13 and 14.

The recirculation system is closed off to the environment, and the system is evacuated to an absolute pressure of −29 in Hg. This vacuum level was held for > 5 minutes to verify that the system was leak tight. Then, NO_2 was delivered to the system from a tank of liquid N_2O_4 and through the surge tank until the system pressure was raised to −14.0 in Hg vacuum, and oxygen was added until the final pressure of the system was 0 in Hg. This resulted in a total system NO_2 partial pressure of ~0.50 atm (absolute) and an NO_2 concentration of 52% by volume in the gas phase.

Tritiated water was introduced to the system as previously described. The tritium trap was charged with a ~10 cm tall bed of silica gel. Initially the tritium trap was valved-out. The iodine trap section was in place but did not contain sorbent. The carrier gas was recirculated at a rate of ~0.25 L/min through the empty iodine bed for 30 minutes. During initial recirculation of the NO_2 gas stream, the pressure downstream of the recirculation gas pump exceeded 20 psi. This was determined to be the result of throttling of the discharge valve from the pump, V-25, combined with the pressure drop across the inlet and outlet valves (V-5 and V-06A), which are part of the Parr reactor. This valving configuration and high discharge pressure exceeded the pressure rating of the M&C TechGroup MP06 pump's Teflon heads and potentially resulted in a leak in the pump head. By reconfiguring the valve lineup to take the primary pressure drop across valve V-08 such that the total flow was reduced to ~0.2 L/min based on the in-line rotameter, the pressure at the discharge could be reduced to under 5 psig and the leak minimized. This initial over-pressurization observed in test ATPT19-5 was not noted in the previous test, ATPT19-4, because the pressure gauge was valved out. If it did occur, it could have affected the result from test ATPT19-4 due to an undetected loss tritiated water. This would explain the higher tritium recovery in test ATPT19-5.

At the end of the 30-minute loading period, the empty iodine trap was valved-out, and the carrier gas was redirected through the silica gel column, while it was cooling from to 40 °C to 33 °C, for 30 minutes in tritium capture mode. The tritium was then recovered from the silica gel under the reverse flow configuration at 140 °C for ~2.5 hours.

Samples were collected in EG after the sorption phase (5a1 and 5a2) and after the desorption phase (5a3, and 5a4). A small fraction of the tritium (~2.8% penetrated the silica gel bed during the sorption phase. Test ATPT19-5 showed slightly better tritium recovery than test ATPT19-4 (~22% recovered vs. ~4% recovered) as shown in Figure 15. The total tritium recovered in Test ATPT19-5 (~23%) was significantly less than in the once-through test ATPT19-3 (~43%indicating either potential losses from the system during the recirculation phase or hold-up within the closed loop system. The losses from the system are more likely due to the known potential for a leak from the pump head.

4.1.6 Test ATPT19-6: Once-through Iodine Recovery

The objective of test ATPT19-6 was to establish once-through iodine recovery into caustic or EG scrubbers with no sorbent in the system. The empty iodine sorbent column was maintained at 150 °C. The flowsheet for this test is shown in Figure 16. This test was repeated seven times at various conditions, and the results are shown in Figure 17. The first three runs 6a–6c) established iodine recovery in caustic while flowing gas at 0.25 L/min for 30 minutes; however, only 10%–14% of the iodine introduced to the system was recovered. Test 6c also included an EG scrubber, which captured as much iodine as the caustic scrubber after 30 minutes of purging.

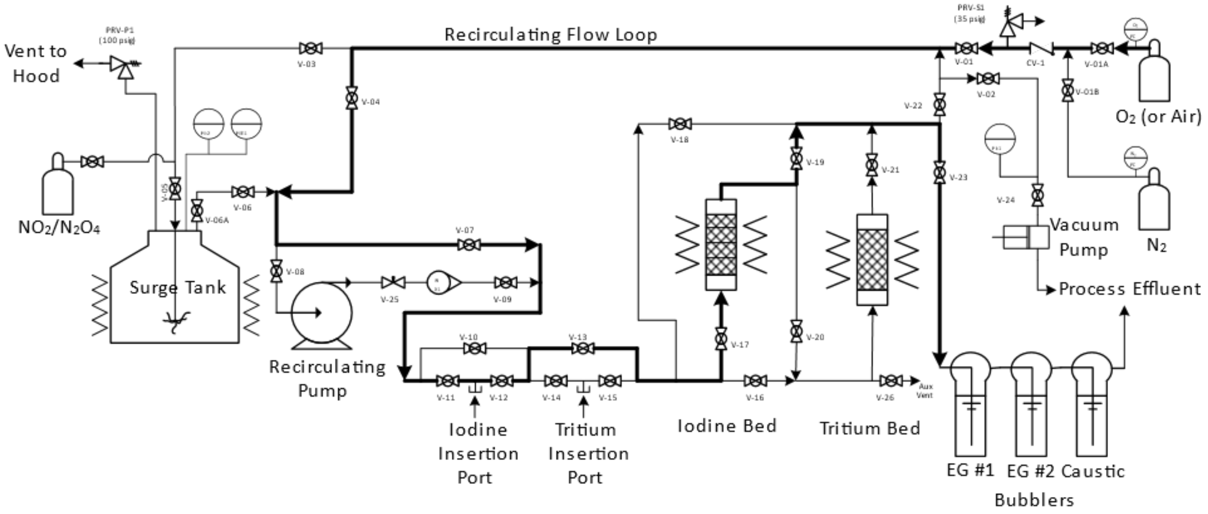

Figure 16: Flow diagram for tests ATPT19-6 and -7 (pass iodine through the trap, with or without AgA).

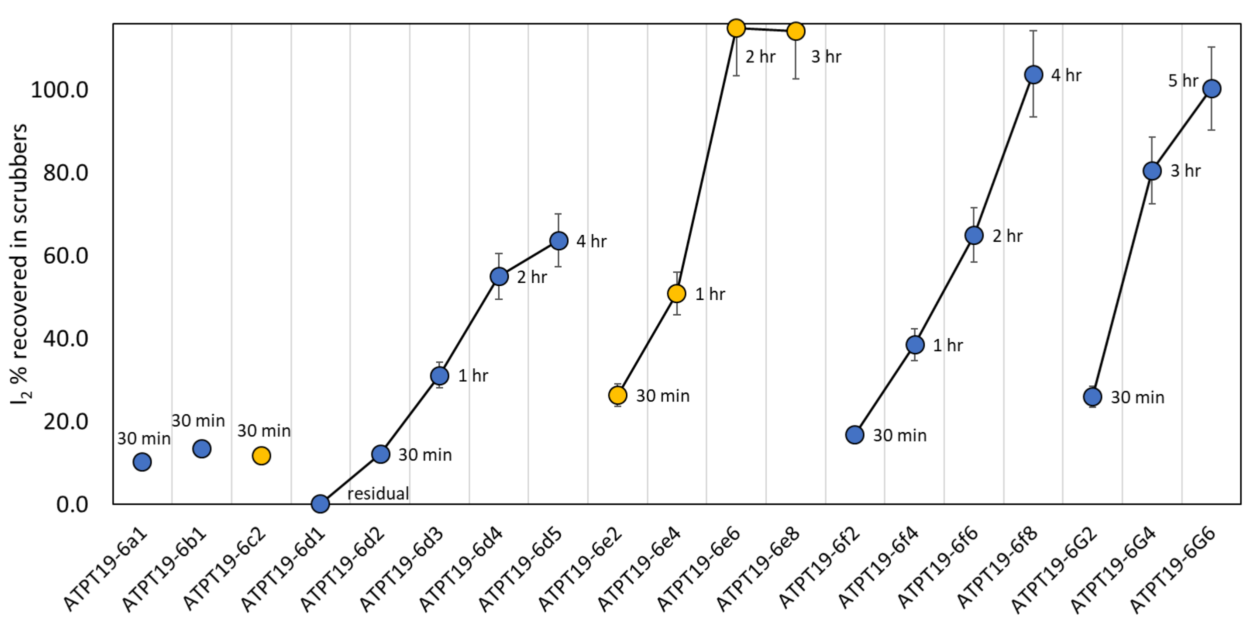

Figure 17: Percent recovery of injected iodine in the scrubbers of tests 6a–6g (once-through with no sorbent in place). Each data point represents the cumulative concentration of multiple scrubbers if more than one was used for that test. The color of the point corresponds to the primary scrubbing solution: ethylene glycol samples are in yellow, and NaOH samples are in blue. Lines connect samples collected over time in the same set of scrubbers during a single test sequence (e.g., the 6d series, etc.).

After tests 6a–6c were completed, parts of the Hastelloy test loop were disassembled to check for iodine residue and moisture in the system using white pipe cleaners that were passed through various line segments and valves. Iodine was observed to have primarily deposited within V-23 and between V-23 and V-22. Possible iodine residue was also found above V-21. The columns were free from iodine residue. Fines from the AgA during test 3c were observed at the junction of V-16 and V-20. Some moisture was observed in the lines near V-23. This liquid was at least partially the result of a process upset resulting from cooling the system prior to opening the connections to the scrubber, resulting in a partial vacuum (due to gas contraction) that pulled some EG from the scrubber back into the line leading to V-23 during the sampling of the test 6c scrubber solution.

Test 6d was similar to test 6c with the empty iodine sorbent column at 150 °C and an O_2 flowrate of 0.25 L/min, but ran for a duration of 4 hours. Test 6e used higher temperatures as well as higher O_2 flowrates (200 °C and 0.5 L/min) for 3 hours. Caustic was the primary scrubbing solution in test 6d and ethylene glycol was used in 6e. The EG turned visibly darker orange over the sampling period of test 6e. Iodine recovery in test 6d was lower than for test 6e. The results are suspect due to potential analytical issues identified by the analytical lab for that specific set of tests. Test 6e shows 100% iodine recovery was achieved in 2–3 hours.

After completion of the combined tests (ATPT19-11a, ATPT19-13a and ATPT19-15a) discussed below, two additional once-through iodine tests were conducted to understand the limited iodine recovery noted in tests 6a–6e. In test 6f, the system was re-piped downstream of V-15/V-13, with Teflon tubing installed and routed directly from the tee to the scrubbers (see Figure 18). Iodine was then introduced into the insertion port and a flowing O_2 stream (0.5 L/min) was used to transport the iodine directly out to the scrubbers in an attempt to capture all of the iodine introduced. Within 30 minutes of the start of iodine flow a black coating of what is believed to be desublimed iodine formed at the junction of the clear Teflon line and the tee, and remained there for at least 2 hours. At the end of a 4-hour purge time, the coating was gone.

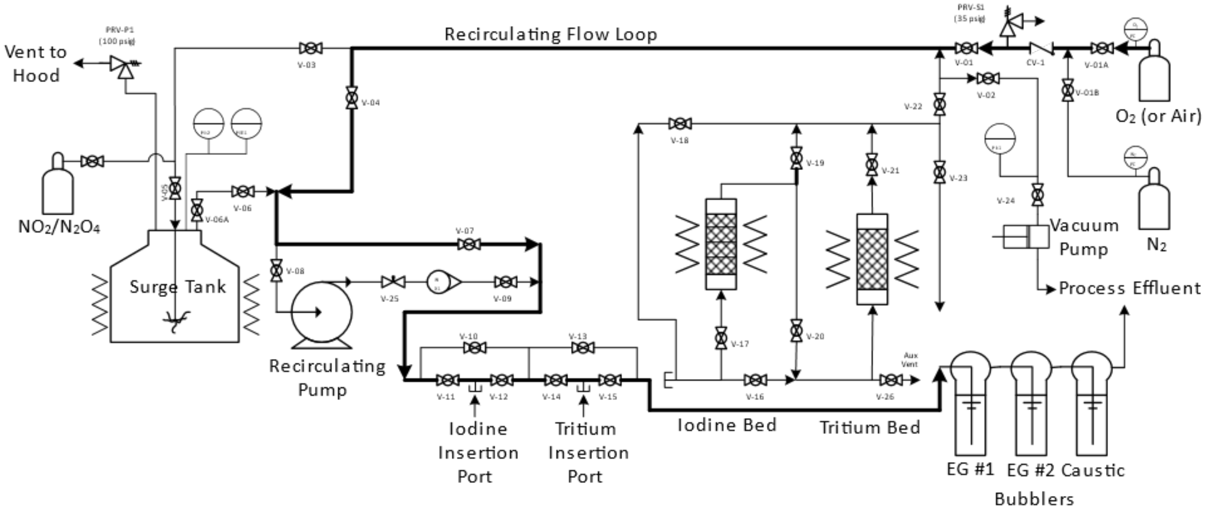

Figure 18: Flow diagram for Test ATPT19-6f (short once-through directly from iodine insertion port to two caustic scrubbers).

The 30-minute sample from test 6f was consistent with the previous tests (6a–6c). A relatively small fraction of the total iodine injected into the system (17%) was recovered after 30 minutes of flow. After 4 hours, 104% of the iodine introduced into the system was recovered based on the analysis of the caustic scrubber solution. These results suggest that the low iodine recovery observed in tests 6a, 6b, and 6c was a result of an insufficient transport time and of iodine desublimation followed by a slower resublimation from the unheated tubing sections used to connect the test system with the bubblers.

Test 6g was also designed to address the apparent loss of iodine in the system and suspected analytical issues associated with iodine analysis in the EG matrix that were later resolved. This system test was conducted using the same configuration as in tests 6a–6e and the carrier gas delivered the iodine into two caustic scrubbers. The iodine was carried with an oxygen flow of 0.25 L/min through the column at 150 °C and into the scrubbers for 3 hours, then the flow rate was raised to 0.5 L/min for an additional 2 hours (for a total of 5 hours of flow time).

In test 6g, 26% of the iodine was recovered after 30 minutes; after 3 hours 80% of the iodine was recovered, and at the end of the 5-hour gas purge 100.4% of the iodine was recovered. These results demonstrate that all iodine introduced into the system can be recovered from the system when no sorbent is present, but this required far longer (3 to 5 hours) than anticipated in any of the initial tests. Full iodine recovery indicated that no iodine was consumed by reaction with metal system components nor did any remain unevaporated within the system The longer transport times are easily explained by the existence of cold spots in the system and an analysis of the iodine vapor pressure and the quantity of iodine saturated air required to transport the quantity of iodine introduced into the system. For example, in test 6a 0.1339 g I_2 was introduced. Based on the iodine vapor pressure, 1.775 mol of air, or 24.77 liters, would be required if saturated with iodine at 21 °C. At a flowrate of 0.25 L/min this would require ~180 minutes (3 hrs) of operation.

4.1.7 Test ATPT19-7: Once-through Iodine Sorption/Desorption by AgA

The objective of Test ATPT19-7 was to demonstrate the quantitative recovery of the injected and adsorbed iodine on the AgA sorbent media. The flowsheet for this test is shown in Figure 16 with the exception that the iodine column or bed was loaded with four AgA sections of ~2.5 cm each. The tritium column was in place but did not contain sorbent. Oxygen was introduced to the system as a carrier gas at 0.25 L/min, and

0.1295 g of iodine was loaded into the iodine insertion port. The flow of O_2 was then directed through the iodine injection segment, and then through the AgA column operated at 150 °C for 30 minutes before discharging into a caustic scrubber. The scrubber was sampled after 30 minutes (sample 7a1) and 2.5 hours (7a2). Approximately 0.05 g or ~36% of iodine introduced was unexpectedly found in the scrubber after 30 minutes and this concentration remained constant over the duration of the test. Within the first 30 minutes, ~3 mL of liquid was observed in the Teflon line coming out of V-23. The origin of the liquid, which is thought to be water, was attributed to the dehydration of the AgA as the bed was heated to its operating temperature. The observed liquid was a condensate that formed at the transition from the heated Hastelloy to the room-temperature Teflon tubing. This liquid likely picked up residual iodine deposits from prior tests which could explain the unexpectedly high iodine concentration in the scrubbers.

Following the run, the AgA was observed to have turned mostly white and a ~1–2 cm band of yellow AgA was observed at the base of the column. The stainless-steel mesh screens that separated the AgA sections were visually examined after the run and were found to be free from corrosion. NAA results show that 112% of the iodine introduced to the system was adsorbed onto the AgA. Virtually all the iodine was contained within the first 2.5 cm of the column. There are several potential sources for the slight excess iodine observed on the AgA. The first is the extent to which the AgA decreased in mass due to water loss during heating and readsorption of water that could impact the weight basis used to determine the iodine loading in terms of weight percent. During simple weighing and drying operations AgA has been noted to adsorb and desorb water with percentage levels change within minutes. Thermogravimetric analysis evaluations of the dehydration of the AgA at operating temperatures indicate that the loaded sorbent would undergo a weight loss of approximately 4 wt%. This weight loss was incorporated into the final calculated iodine recovery estimates for this and all following tests. The second source of the error is that associated with the NAA measurements themselves. Based on the data from MURR, these uncertainties appear to be between ~1% and 3%. The third part is an estimate of the variability of a single ~0.2 g subsample from the larger 5–8 g sample. (The subsamples in this case were on the order of 0.3–0.4 g.) The variability with the ~0.2g subsamples was previously determined to be ~12%–27% for subsamples that represented 2%–3% of a large deep bed and ~10% for subsamples that represent significantly larger fractions of thin beds (Jubin et al. 2017b). For this estimation of sampling variance, the value of ~20% was assumed as similar subsampling variability expected here. As a result, the once-through test appears to be within the expected experimental variability, and all the iodine introduced into the system can be recovered on the AgA.

4.1.8 Tests ATPT19-8–10: Iodine Only with and without Recirculation

The preliminary data obtained on the tritium hold-up within the system showed considerable water hold-up would occur on the AgA and that only a limited recovery of the tritium from the silica gel was obtained at the levels contained in the test system. Based on these observations it was determined that rather than conduct strictly iodine only tests (i.e., tests 8–10), the same information could be determined from conducting the combined iodine / tritium tests designated as tests ATPT19-11, -13, and -15. The major variant from the plan was the elimination of the silica gel from the test loop. The tests with tritium alone showed that the silica gel would capture the tritiated water, but quantitative recovery was difficult. This may be because the silica gel was not near saturation with tritiated water. In any case, it was safe to assume that the silica gel would, if it were in place, adsorb the tritiated water that would pass through the AgA bed.

4.1.9 Test ATPT19-11: Once-through Iodine and Tritium Sorption/Regeneration by AgA

The objective of test ATPT19-11 was to demonstrate the quantitative recovery of once-through iodine and tritium adsorption, and recovery of any tritium co-absorbed by the AgA. This could be considered a standard air TPT off-gas system without an additional silica gel trap. The surge tank represents the voloxidizer. Oxygen was used as the carrier gas. The flowsheet for this test is shown in Figure 19.

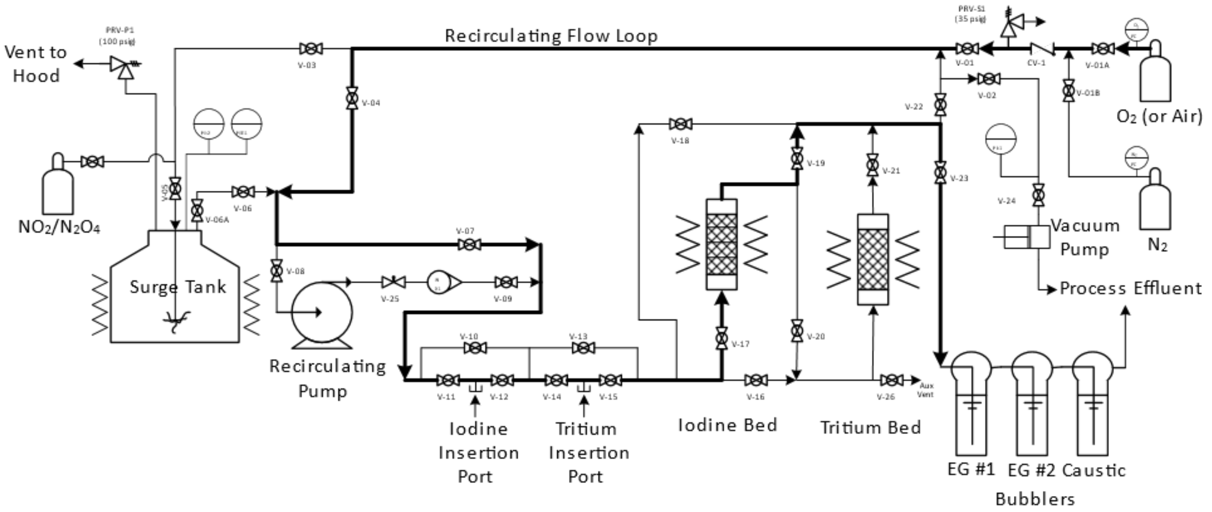

Figure 19: Flow diagram for Test ATPT19-11 (once-through iodine and tritium onto AgA).

Three sections of AgA were loaded into the iodine trap and separated by glass wool and steel mesh. The lower section was 1.4 cm tall, the middle section was 2.3 cm tall, and the top section was 6.3 cm tall. The AgA was dried for 3 hours at 200 °C before the run, and the tritium trap was empty.

At the start of this test, 0.1349 g of iodine and 100 µL of tritiated water were introduced to the system through the injections ports, and the system was brought to operating temperature with a flowing gas stream bypassing the injection point. At time zero, the flow was routed through the charging sections and continued through the to the 150 °C AgA trap at 0.25 L/min. The gas stream exiting the system was routed to an EG scrubber followed by a caustic scrubber, and both scrubbers were sampled at 30 minutes (11a1, 11a2), 1 hour (11a3, 11a4), and 2 hours (11a5, 11a6). After 2 hours operating in the TC mode, the temperature of the AgA bed was raised to 250 °C, and the flow rate was increased to 0.5 L/min to desorb tritium and any physically adsorbed iodine for 4 hours (2 hour samples: 11a7 and 11a8; 4 hour samples: 11a9 and 11a10).

The tritium activity was below 1.0E+00 Bq/mL (0.01% expected activity) in all of the scrubber samples taken during the adsorption phase of the test (11a1–11a6) and was only slightly elevated (<3.0E+01 Bq/mL or 0.3% expected activity) in the tritium desorption phase scrubber solution samples. This would appear to demonstrate that the tritium is effectively adsorbed to the AgA under these operating conditions and that it is not recovered from the AgA at a regeneration temperature of 200 °C. Samples from the AgA bed were analyzed for gross tritium content by placing a small portion of the material in a vial of scintillation solution, and after allowing time for some degree of exchange to occur, the solution was counted to determine the resulting activity. At best, this is a gross estimation of the residual tritium. However, the results indicate that a measurable amount of tritium was in the AgA (~6.5% of the amount introduced), and ~60% of the tritium found was contained in the first segment of the bed.

The iodine concentrations detected in all the scrubber samples were below 5.0E+00 µg/mL (<1% of iodine introduced) (Figure 20).

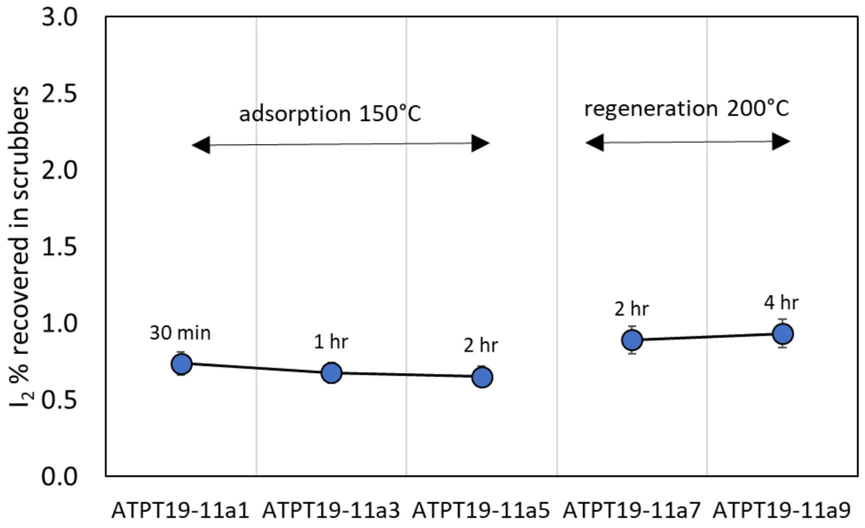

Figure 20: Percentage of iodine recovered, relative to iodine introduced to the system, in scrubbers from test ATPT19-11 (once-through adsorption onto AgA). The data points represent the cumulative iodine recovered between two scrubbers. Lines connect samples collected over time in the same set of scrubbers during a single test sequence.

Upon the removal of the AgA from the column, the first or inlet segment (~1 cm) had a yellow coloration believed to be indicative of iodine adsorption. The NAA results for this test indicate that 114% of the iodine introduced into the system was recovered on the AgA with ~90% loaded in the first bed segment (1.4 cm). This value of 114% is within the expected values for quantitative iodine recovery based on subsampling of the bed segments and weight loss caused by drying of the AgA. Overall, the data indicate that iodine is completely removed from the system with AgA.

4.1.10 Test ATPT19-13: Recirculation of Iodine and Tritium Sorption/Regeneration by AgA with O_2

The objective of test ATPT19-13 was to demonstrate the quantitative recovery of iodine and tritium during O_2 recirculation with the surge tank in place. This is a variant of the standard air TPT off-gas system but increases the sorbate concentrations by recirculating O_2. The surge tank represents the voloxidizer. The flowsheets for this test are shown in Figures 21 and 22.

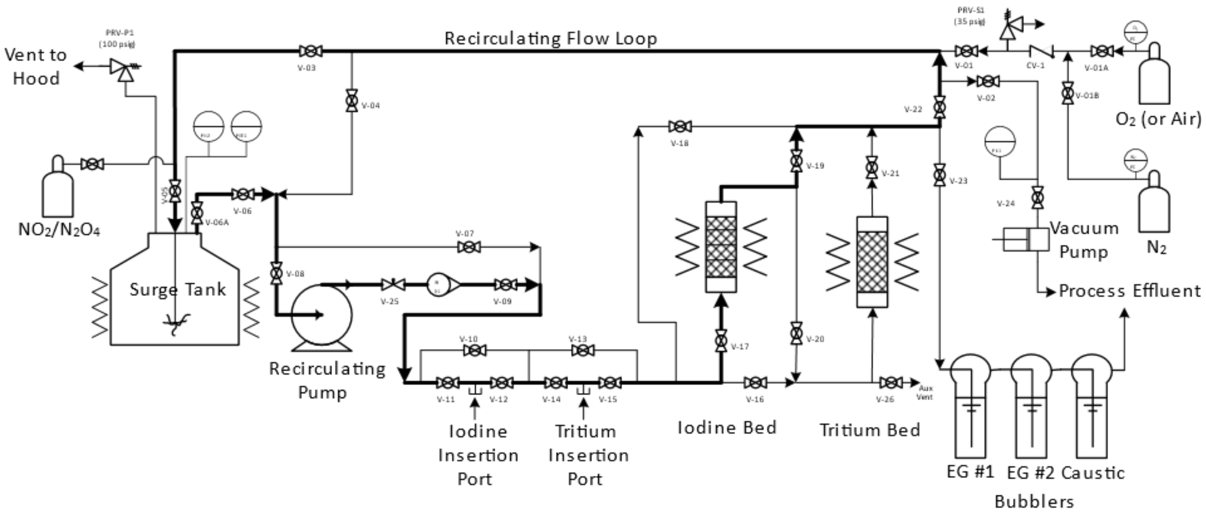

Figure 21: Flow diagram for tests ATPT19-13 and -15 part 1 (recirculation through AgA).

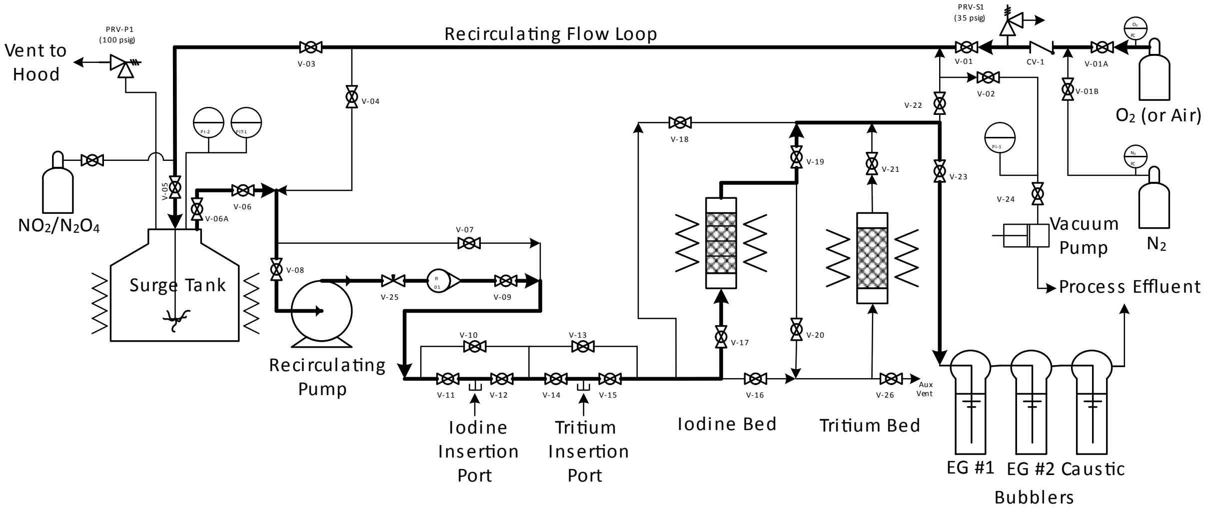

Figure 22: Flow diagram for tests ATPT19-13 and -15 part 2 (desorption of AgA to scrubbers).

Three sections of AgA were loaded into the iodine trap and separated by glass wool and steel mesh: the lower section was 1.6 cm tall, the middle section was 2.3 cm tall, and top section 5.9 cm tall. The AgA was dried under O_2 at 200 °C for 2 hours before the start of the run. The tritium trap was empty. The system was purged for 30 minutes to establish flow through the surge tank. Then the system was closed to the atmosphere and iodine and tritiated water charging chambers were valved-in to a recirculation loop for 2 hours with O_2 flowing at ~0.25 L/min, passing through the AgA column which was maintained at 150 °C. The AgA column was then isolated and the system was vented to an ethylene glycol and caustic scrubber for 30 min to remove any sorbate that did not adhere to the column (sample 13a1,13a2). Finally, the AgA column was valved-in to recover tritium from the AgA by purging it at 200 °C for over 4 hours (samples 13a3–13a6).

Iodine concentrations were below 1.0E+00 µg/mL in all scrubbers (<0.1% iodine introduced) (see Figure 23). Similar to previous tests, the bottom ~ 1 cm of the AgA beads had a yellow coloring believed to be indicative of iodine adsorption. The stainless-steel mesh screens that separated the AgA sections were visually examined after the run and were found to be free from corrosion. The NAA results show that 132% of the iodine introduced into the system was recovered on the AgA with >99% of the iodine in the first segment. This value is above the expected amount and significantly higher than the yields for tests 7a and 11a. This may reflect recovery of residual iodine deposits in the lines connecting the surge tank to the rest of the system, as many of these lines could not be insulated well. Nevertheless, <1% recovery in the scrubbers and >100% onto the AgA shows that iodine can be completely removed from the system at these test conditions.

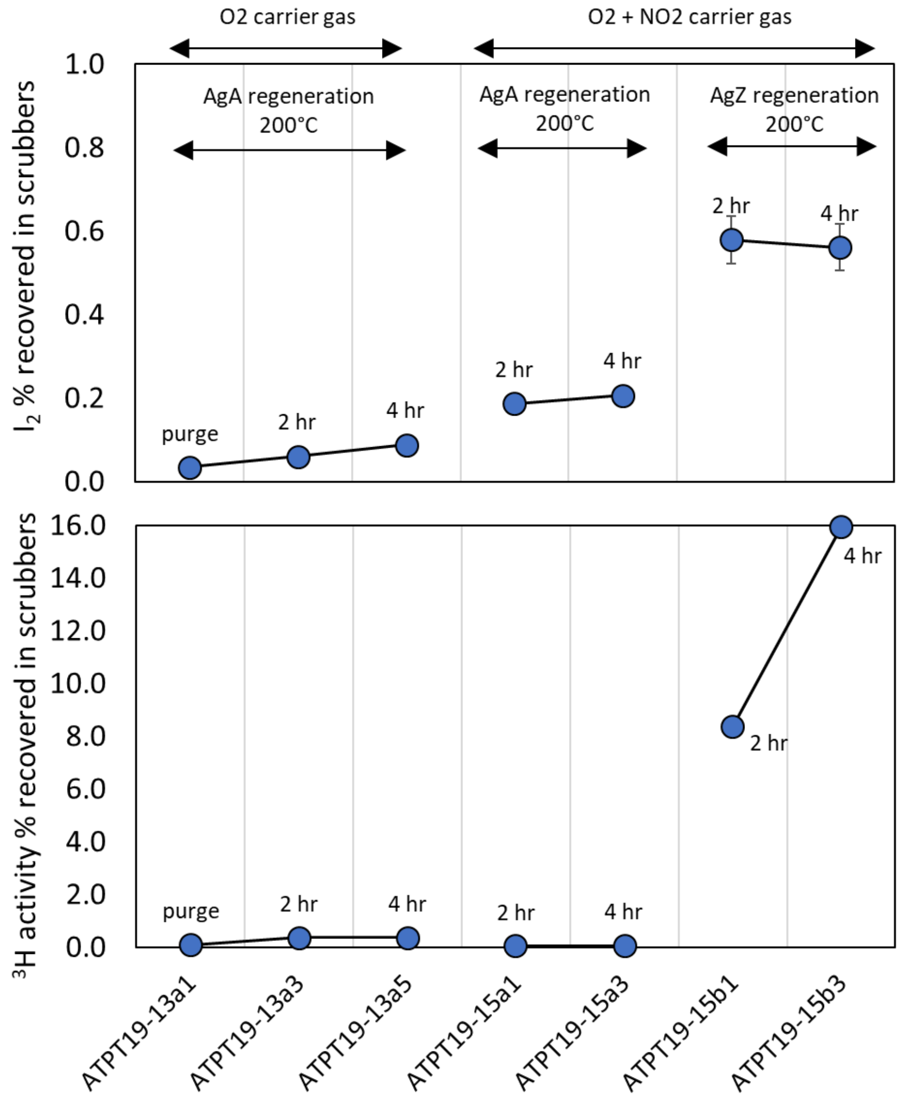

Figure 23: Percentage of the tritium and iodine recovered in the scrubbers in tests 13a (recirculation with O_2 carrier gas) and 15a and b (recirculation with NO_2 + O_2 carrier gas). Data points represent the cumulative activity or concentration in both scrubbers when two scrubbers were used. Lines connect samples collected over time in the same set of scrubbers during a single test sequence. The sorbents were AgA for Tests 13a and 15a, and AgZ for test 15b.

Tritium activity was below 4.0E+01 Bq/mL, or approximately 0.4% of the expected tritium activity, in all the scrubber solutions. Samples from the AgA bed were analyzed in a similar manner to those from tests 11a for gross tritium content. As with the results from test 11a, the analysis indicates that a measurable amount of the tritium introduced into the system was retained on the AgA (~8.4% of the amount introduced) and that ~80% of that tritium was found in the first segment of the bed.

4.1.11 Test ATPT19-15: Recirculation of Iodine and Tritium Sorption/Regeneration by AgA or AgZ with NO_2

The objective of test ATPT19-15a was to demonstrate the quantitative recovery of iodine and tritium during NO_2 recirculation with the surge tank in place. The flowsheet for this test is shown in Figures 21 and 22. Three sections of AgA were loaded into the iodine trap and separated by glass wool and steel mesh: the lower section was 1.4 cm tall, the middle section was 2.4 cm tall, and the top section was 6.0 cm tall. The AgA was dried under O_2 at 200 °C for 2 hours before the start of the run. The tritium trap was empty.

A vacuum was pulled on the system which was then filled with approximately equal portions of NO_2 and O_2. The iodine and tritiated water were introduced to a recirculation loop for 2 hours at a gas flow rate of ~ 0.25 L/min, passing through the AgA column at 150 °C. Then the AgA column temperature was raised to 200 °C and "regenerated" by purging with O_2 at 0.5 L/min for 4 hours into EG and caustic scrubbers in series (samples 15a1–15a4).

The iodine concentration (see Figure 23) in the scrubbers during regeneration is slightly higher than in test 13a (O_2 carrier gas) but still below 1.0E+00 µg/mL (<0.2% of iodine introduced.). The NAA results show 102% of the iodine introduced to the system was recovered on the AgA with >99% of the iodine found in the first segment of the bed. Again, it appears that the AgA completely removes iodine carried by a mixture of NO_2 and O_2 from a recirculating system.

The tritium activity measured in the scrubbers after regeneration was 5.1 E+00 Bq/mL, or < 0.1% of the introduced tritium activity, again showing that tritium cannot be removed from the AgA at these test conditions. Analysis of samples from the AgA bed for residual gross tritium content also show that a portion of the tritium introduced into the system was retained on the AgA (~4.8% of the amount introduced) and that >50% of that tritium was found in the first segment of the bed.

Test 15b was a repeat of 15a except that AgZ was used in place of the AgA. All other test conditions were kept the same. Slightly more of the iodine that was introduced to the system was recovered in the scrubbers at the end of the 4-hour sampling period but still <0.6%. The NAA results show that 123% of the iodine introduced into the system was recovered on the AgZ. However, the iodine penetrated deeper into the AgZ than into the AgA. Most (82%) of the iodine was in the first 1.3 cm of AgZ, and 17% was in the next 1.2 cm segment of AgZ. Like the AgA, the AgZ removed all the iodine introduced into a recirculating system and carried by a mixture of NO_2 and O_2. The tritium activity measured in the scrubbers after the tritium purge was 1.47E+03 Bq/mL, or approximately 16% of the expected tritium activity (Figure 23). Based on this single test, recovering the tritium from AgZ as separate stream appears easier than recovery from AgA, but a longer and potentially higher temperature desorption period than used in this test would be required. Analysis of samples from the AgZ bed for residual gross tritium content shows that a measurable portion of the tritium introduced into the system was retained on the AgZ (~7.9% of the amount introduced), but the tritium distribution was shifted to the discharge segment of the bed (73% of the tritium was observed in the final segment), potentially indicating that the desorption process was proceeding and should have run longer.

5. CONCLUSIONS

A new test bed for the advanced TPT off-gas system that addresses the issues identified in the fiscal years 2017 and 2018 scoping tests was designed. The test bed used high nickel alloys wherever possible to address the previously observed corrosion issues, a redesigned surge tank to permit ease of purging, an improved feed system, and a more modular system design that allowed testing of incremental increases in system complexity to allow the isolation of any element that results in changes to recovery of target species. No visible corrosion was observed in the system.

Accompanying that design, a multifaceted test plan was completed. A total of 22 tests were completed: 10 tests examined tritium behavior under varying operating parameters, 8 tests examined iodine behavior under varying operating parameters, and 4 tests examined both tritium and iodine behavior. The results from these 22 tests are summarized in Table 3. While the separate recovery of tritium proved problematic, the series of tests were viewed as successful. Based on the analytical data obtained from these tests and the discussions in Section 4, the following conclusions can be drawn:

- With Hastelloy components comprising most of the test system, full recovery is achieved for both iodine and tritium sorbates when no sorbent is in place, indicating little to no loss of sorbates to system corrosion. Approximately 96% of the tritium could be recovered from the empty test rack in 2 hours. Approximately 100% of iodine can be recovered from the empty test rack in 3–5 hours.
- Tritium adsorbs to, and is not recovered from, a 10 cm column of silica gel, when the silica gel is dried at 90–200 °C, and subsequently tritium is loaded between 30 and 40 °C and the silica gel regenerated between 90 and 140 °C. Regenerating the column in a reverse flow pattern permitted up to 43% of the tritium to be recovered in 3 hours. Recovery is lower (~23%) when tritium is first recirculated through the system before being adsorbed to the silica gel.
- Tritium adsorbs to, and is not recovered from, a 10 cm column of AgA when the AgA is first dehydrated at 200 °C, tritium is loaded at 150 °C and the AgA then regenerated at 200–250 °C. Under these conditions, less than 2% of the initially injected tritium was recovered in the scrubbers. This was confirmed by four tests (3c, 11a, 13a, and 15a). Tritium presence on the AgA is confirmed with scintillation counting of the AgA media, however these results are mostly qualitative.
- Co-adsorbed tritium is more easily recovered from Ag-mordenite (AgZ) than from AgA. After loading tritium onto the AgZ at 150 °C, approximately 16% was recovered during the tritium desorption phase at 200 °C versus <0.4% for AgA desorbed at 200 °C
- Iodine takes significantly longer (3–5 hours) than tritium to be recovered from the test system. This may be due to repeated iodine desublimation and resublimation in portions of the system that could not be heat traced (e.g., the Teflon lines leading to the bubblers themselves). Vapor pressure calculations indicate that 44 L of O_2 would be required to transport the quantity of iodine in the system at 18 °C; therefore, 3 hours of online flow time at 0.25 L/min should be sufficient to transport all iodine out of the system, but we find that a complete iodine purge takes closer to 5 hours. This can be caused by desublimation in cool or dead zones of piping where subsequent resublimation must occur. In dead zones this may be further limited by diffusional transport.

Table 3: Summary of test results.

					Sorption Conditions			Desorption Conditions				Sorption losses		Desorption Recovery		
Test ID	Sorbent (Iodine/ Tritium)	Mode of Operation	Carrier Gas	Sorbate	Transfer Line / Adorption Bed Temp	Gas Flow Rate in (L/min)	Sorption Duration (min)	Desorption Temp	Gas Flow Rate (L/min)	Desorption Duration (min)	Bubbler Solution(s)	Tritium (%)	Iodine (%)	Tritium (%)	Iodine (%)	Retained on Iodine Sorbent (%)
Tritium only tests																
ATPT19-1a	None/None	Once-through / no sorbent	O_2	tritium	40°C	0.25	30	N/A	N/A	--	EG + NaOH	N/A	--	51.7	--	--
ATPT19-1b	None/None	Once-through / no sorbent	O_2	tritium	40°C	0.25	30	N/A	N/A	--	EG + NaOH	N/A	--	86.1	--	--
ATPT19-1c	None/None	Once-through / no sorbent	O_2	tritium	40°C	0.25	30	N/A	N/A	--	EG + NaOH	N/A	--	88.2		--
ATPT19-1d	None/None	Once-through / no sorbent	O_2	tritium	75°C	0.25	120	N/A	N/A	--	EG	N/A	--	84.1	--	--
ATPT19-2a	None/Silica gel	Once-through w/ sorbent	O_2	tritium	40°C	0.25	30	90°C	0.5	120	EG + NaOH	18.3	--	0.4	--	--
ATPT19-3a	None/Silica gel	Recirculation 30 min	O_2	tritium	40°C	0.25	20	90°C	0.25	120	EG + NaOH		--	0.1	--	--
ATPT19-3b	None/Silica gel	Desorption #2 Reverse flow	O_2	tritium	N/A	N/A	--	140°C	0.25	180	EG + NaOH		--	43.0	--	--
ATPT19-3c	AgA/None	Recirculation 30 min	O_2	tritium	150°C	0.25	30	200°C	0.25	80	EG + NaOH		--	1.4	--	--
ATPT19-4a	None/Silica gel	Recirculation w/ Surge Tank	O_2	tritium	30°C	0.25	30	140°C	0.25	~150	EG		--	3.6	--	--
ATPT19-5a	None/Silica gel	Recirculation w/ Surge Tank	NO_2	tritium	40–33°C	0.25	33	140°C	0.25	~150	EG + NaOH		--	22.6	--	--
Iodine only tests																
ATPT19-6a	None/None	Once-through / no sorbent	O_2	iodine	150°C	0.25	30	N/A	N/A	--	NaOH	--	N/A	--	10.2	
ATPT19-6b	None/None	Once-through / no sorbent	O_2	iodine	150°C	0.25	30	N/A	N/A	--	NaOH	--	N/A	--	13.4	
ATPT19-6c	None/None	Once-through / no sorbent	O_2	iodine	150°C	0.25	30	N/A	N/A	--	EG + NaOH	--	N/A	--	11.6	
ATPT19-6d	None/None	Once-through / no sorbent	O_2	iodine	150°C	0.25	240	N/A	N/A	--	NaOH	--	N/A	--	63.7	
ATPT19-6e	None/None	Once-through / no sorbent	O_2	iodine	200°C	0.5	180	N/A	N/A	--	EG + NaOH	--	N/A	--	114.9	

					Sorption Conditions			Desorption Conditions				Sorption losses		Desorption Recovery		
Test ID	Sorbent (Iodine/ Tritium)	Mode of Operation	Carrier Gas	Sorbate	Transfer Line / Adsorption Bed Temp	Gas Flow Rate in (L/min)	Sorption Duration (min)	Desorption Temp	Gas Flow Rate (L/min)	Desorption Duration (min)	Bubbler Solution(s)	Tritium (%)	Iodine (%)	Tritium (%)	Iodine (%)	Retained on Iodine Sorbent (%)
ATPT19-6f	None/None	Once-through / no sorbent	O_2	iodine	NA	0.5	240	N/A	N/A	--	NaOH	--	N/A	--	103.8	
ATPT19-6g	None/None	Once-through / no sorbent	O_2	iodine	150°C	0.25	300	N/A	N/A	--	NaOH	--	N/A	--	100.4	
ATPT19-7a	AgA/None	Once-through w/ sorbent	O_2	iodine	150°C	0.25	30	N/A	N/A	150	NaOH	--		--	38.9[a]	111.8
Combined Iodine/Tritium tests																
ATPT19-11a	AgA/None	Once-through	O_2	iodine + tritium	150°C	0.25	120	250°C	0.25—0.5	240	EG + NaOH			0.3	0.9	113.5
ATPT19-13a	AgA/None	Recirculation w/ Surge Tank	O_2	iodine + tritium	150°C	0.25	120	200°C	0.5	223	EG + NaOH			0.4	0.1	131.8
ATPT19-15a	AgA/None	Recirculation w/ Surge Tank	NO_2	iodine + tritium	150°C	0.25	120	200°C	0.5	240	NaOH			0.1	0.2	102.4
ATPT19-15b	AgZ/None	Recirculation w/ Surge Tank	NO_2	iodine + tritium	150°C	0.25	120	200°C	0.5	240	NaOH			15.9	0.6	122.9

Notes:

a. Suspected origin of this iodine was from residual iodine deposits in the lines. NAA results of the AgA indicate all iodine injected was captured by AgA.

- The integrated tritium and iodine tests (11a, 13a, and 15a) indicate that less than 1% of the iodine introduced into the system passes through the AgA bed in either the once-through mode or in the desorption mode following iodine loading in recirculation mode. The iodine recovery in the scrubbers ranged from below detection limits (<0.1%) to 0.6% which indicate decontamination factors of 166 to >1000 during the once through IC mode. Further testing with deeper beds would be required to determine if iodine decontamination factors >1000 can be achieved.
- Iodine recovery using AgA sorbent appears effective under all conditions tested: once-through, recirculation with O_2, and recirculation with NO_2+O_2. In all cases the AgA captures and retains ~100% of iodine.
- Silver-mordenite (AgZ) performs to the same degree as AgA in the iodine retention test under recirculation with NO_2+O_2 conditions. However, the iodine penetration into the bed observed in the single test with recirculating NO_2+O_2 appeared to be deeper into the AgZ. Iodine desorption from the AgZ following loading appeared to be slightly higher than for AgA.
- Analysis of the iodine sorbent beds from the integrated tritium and iodine tests (11a, 13a, and 15a) indicate quantitative iodine recovery (within anticipated experimental errors) can be achieved.
- This system may provide a means for the quantification of the iodine released by the ATPT process within the experimental uncertainties. This would support 0.33 to 1.00 kg demonstrations of the ATPT process with actual used nuclear fuel.
- Three sources of experimental errors were identified: (1) variation in the actual AgA weight basis for the iodine mass percent due to water adsorption/desorption; (2) NAA analytical uncertainty; and (3) uncertainty associated with the bed segment subsampling.
- Based on the tritium adsorption behavior of the AgA, this material does not appear to be suitable for a system if separate tritium recovery is desired. Potentially a different form of alumina is required or the bed must be heated to a higher temperature to release the tritiated water.
- Examination of the internals of the test system at selected points showed no signs of the corrosion that had been observed in the previous set of tests.

Follow-on tests of this system should be conducted to demonstrate the separate recovery of iodine and tritium from tests such as those performed in ATPT19-11a, -13a, and -15b. However, the following changes to the test bed and test conditions should be made:

1. Replace the recirculation pump with one that does not use a split Teflon head
2. Use AgZ rather than AgA or another support substrate for the $AgNO_3$.
3. Increase the desorption time (increasing the desorption temperature is currently limited by the Hastelloy valve seats and packing.)
4. Increase the tritium desorption time for the silica gel bed or flow in the reverse direction to that used during adsorption phase.
5. Explore methods to reduce the experimental uncertainties especially these associated with iodine measurement. This should focus decreasing the ±10%analytical uncertainty and the 10+% uncertainties associated with the subsampling of the sorbent beds for NAA.

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