COMPARISON OF EXTENDED AND ACCELERATED VOG TESTS

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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 ^{129}I. The dissolver off-gas and more dilute vessel off-gas streams (VOG) must both be targeted when mitigating ^{129}I environmental release because each contains some amount of iodine. Iodine-129 will likely be found as elemental iodine (I_2) and methyl iodide (CH_3I) in these off-gas streams. Reduced silver-exchanged mordenite (AgZ or Ag^0Z) has been investigated as a potential sorbent for iodine abatement and is studied here under prototypic VOG conditions.

Because of the relatively low iodine abundance and high flow rates of the VOG, total iodine concentrations are expected to be in the parts per billion range. Thus, VOG experiments need to run for extended durations at low concentrations to reach sorbent saturation. Because the sorbent will be exposed to oxidizing gas streams for extended periods of time, aging effects and sorbent degradation need to be considered when designing a sorbent-based abatement system.

Three sets of experiments were performed with the aim of determining how the adsorption of iodine by AgZ is affected by differing test durations and gas compositions. The first tested the capacity of sorbent aged for 8 months under a humid air stream. The second tested differences in sorbent capacity and mass transfer zone (MTZ) length during high-concentration (1200 ppb_v) CH_3I loading in a humid nitrogen gas (N_2) stream and a humid air gas stream over 28 days. The third tested sorbent capacity and MTZ length during low concentration (< 200 ppb_v) CH_3I and I_2 loading in a humid air stream over 9 months.

The results of these tests suggest that AgZ capacity drops by ~50% after 1 month of aging, by ~60% after 2 months of aging, and then does not continue to decrease significantly at up to 8 months of aging.

One-month aging tests that compared N_2 and air as the gas stream diluent resulted in similar maximum loading capacities and overall loading curves, indicating that the effects of aging cannot be mitigated by avoiding air as the balance gas.

The 9-month extended VOG tests did not result in clear sorbent saturation at the inlet, but it can be inferred using an assumption of a maximum capacity of 45 mg I/g AgZ. Applying this assumption, then the MTZ of CH_3I is 12.8 cm, and the MTZ of I_2 is 12.4 cm. These results are similar to previous tests. Comparisons to tests completed at Idaho National Laboratory suggest that the CH_3I MTZ may be dependent on concentration. Future work should re-evaluate the design of the VOG iodine capture system with this updated data and should seek to understand fundamental characteristics of CH_3I adsorption by AgZ, such as the effect of concentration, gas velocity, and gas composition.

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ACRONYMS

AgZ	silver exchanged mordenite
DF	decontamination factor
MTZ	mass transfer zone
NAA	neutron activation analysis
ORNL	Oak Ridge National Laboratory
UNF	used nuclear fuel
VOG	vessel off-gas

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1. INTRODUCTION

US regulations will require the removal of ^{129}I from the off-gas streams of any used nuclear fuel (UNF) reprocessing plant before discharge of the off-gas to the environment. The release of volatile radioactive iodine is governed by three regulations in the United States (40 CFR 61, 40 CFR 190, and 10 CFR 20). These regulations govern both total release limits for iodine and the potential combined dose from the four volatile radionuclides of most concern for aqueous-based UNF reprocessing (^{3}H, ^{14}C, ^{85}Kr, and ^{129}I). The required plant decontamination factor (DF) for iodine will vary based on fuel burnup, cooling time, and other factors but is very likely to be >1000 and could be as high as 8000 (Jubin et al. 2012b). Achieving this level of decontamination will require iodine capture from multiple off-gas streams within an aqueous reprocessing plant, including the vessel off-gas (VOG) stream associated with the solvent extraction step of aqueous reprocessing.

In 2016, an engineering evaluation of an off-gas treatment system for a UNF reprocessing plant was performed, and included within that evaluation was a conceptual design for an iodine abatement system designed to remove iodine from the VOG using reduced silver mordenite (AgZ) (Jubin et al. 2016). The VOG could contain 0.5–2.5% of the total iodine inventory of the processed UNF. As compared with other off-gas streams found within the plant, the VOG will contain a higher proportion of organic iodides (e.g., CH_3I and potentially longer chain organic iodides) at a significantly lower concentration (parts per billion for VOG streams, but other gas streams may contain iodine at parts per million) (Jubin et al. 2013). In design of the conceptual treatment system, two data gaps decreased the fidelity of the projected bed size. The first was the lack of information about the mass transfer zone (MTZ) for iodine adsorption by AgZ under VOG conditions—for the design of the conceptual system it was assumed to be 1 m. The second was that the total iodine loading of AgZ was assumed to be conservatively low (1 wt% or 10 mg I/g AgZ) because of the effects of sorbent aging, the dilute iodine concentration of the stream, and the possibility that adsorption of organic iodides by AgZ will be less efficient than adsorption of elemental iodine. These assumptions resulted in high consumption of the silver-based sorbent (18.3 kg/day) and bed changeout frequencies of 7.23 months.

The 2016 engineering evaluation and a 2015 associated report assessing the knowledge base about organic iodine removal from off-gas streams prompted the initiation of collaborative research by Oak Ridge National Laboratory (ORNL) and Idaho National Laboratory focused on organic iodide adsorption by silver-based sorbents (Bruffey et al. 2015). In 2016, two iodine adsorption tests were performed using AgZ as the adsorbent. The adsorption of both I_2 (at 7 ppb_v) and CH_3I (40 ppb_v) were examined with test parameters representative of VOG conditions. The concentrations of iodine in the feed streams for these tests were some of the lowest reported in open literature at the time. The AgZ sorbent remained below saturation at the conclusion of 12–13 weeks of testing, and I_2 penetrated 2.2 cm into the bed and CH_3I penetrated 2.4 cm into the sorbent bed.

After these initial tests, more expansive experimentation evaluated the effects of concentration on CH_3I adsorption by both AgZ and silver-functionalized silica aerogel (AgAero), an alternative silver-based iodine sorbent in early development (Jubin et al. 2017). Concentrations from 40 to 1000 ppb_v were evaluated and it was found that the adsorption profile in the bed was unchanged over that concentration range. The sorbents were not saturated after testing, and the maximum observed iodine (fed as CH_3I) loading for AgZ was 0.9 wt%. These tests were of varying duration (4–94 days, depending on CH_3I concentration, with the total amount of iodine delivered to the sorbent bed remaining constant), and sorbent capacity loss due to extended exposure to moist air (aging) was not controlled for in these

experiments. Aging of AgZ by moist air has been shown to decrease sorbent capacity by ~50% after 1 month and by ~60% after 4 months (Jubin et al. 2012a). Thus, the effective capacity of 4-month humid-aged AgZ is 40% of fresh AgZ. Assuming 100% silver utilization for unaged AgZ with a silver content of 10 wt%, this translates to a maximum capacity of 4.7 wt% iodine (47 mg I/g AgZ).

To better understand the long-term performance of AgZ as a iodine adsorber in prototypical VOG conditions, an 8-month test was performed in which 50 ppb_v I_2 was loaded onto a deep bed of AgZ under a humid air stream at 150°C (Jubin et al. 2018). The sorbent was not saturated during this test, and iodine penetrated 10.5 cm into the sorbent bed. Using these data and assuming a sorbent saturation level of 45 mg I/g AgZ sorbent (corresponding to 4-month humid aged AgZ), the MTZ for I_2 adsorption by AgZ in VOG conditions was estimated to be 13 cm.

The studies presented in this report build on the body of work described here. First, the iodine capacity of 8-month humid aged AgZ was determined. This extends the time frame examined by previous aging studies and is consistent with the potential frequency that sorbent bed changeout could be required.

Second, an effort was made to mitigate the effects of aging during experimentation through the use of nitrogen as the balance gas rather than dry air that is known to decrease the capacity of AgZ by up to 40% after 4 months of exposure (Jubin et al. 2012a). The ability to limit aging, combined with a lack of observed adsorption effects due to iodine concentration in the feed gas at concentrations up to 1 ppm_v would provide the ability to conduct "accelerated" or high concentration, limited duration testing. Accelerated testing could substantially increase the amount of data that could be obtained during a specified time period, which is currently limited by the need to conduct tests for ≥4 months to capture aging effects.

Third, two extended duration tests (9 months in duration) were performed to characterize the adsorption of CH_3I and I_2 by AgZ in VOG conditions. These tests extend previous work to CH_3I, which is expected to be a predominant species in the VOG, and allow for further development of MTZ estimations. Together, the data returned from this testing can be used to refine the design of the VOG iodine abatement system provided by Jubin et al. (2016).

2. MATERIALS AND METHODS

Silver mordenite (AgZ) containing 9.4 wt% silver was obtained from Molecular Products in an engineered pelletized form (Ionex-Type Ag 900 E16). Before use in testing, the sorbent material was reduced by exposure to a 4% H_2 blend in Ar at 270°C for 10 days. After reduction, the material was stored under argon to limit oxidation by air. Details about this procedure are provided by Anderson et al. (2012).

Owing to the corrosive nature of iodine, especially in the presence of water, the materials of construction for the system were carefully selected to minimize iodine loss to system components and piping. The sorbent beds were contained within glass columns (internal diameter = 2.73 cm) and separated by glass frits. The humid air and iodine supply streams were piped through separate lines of 316 stainless steel tubing. The two streams were blended together in a glass tee directly before introduction into the sorbent bed.

After iodine loading, the iodine content of loaded AgZ was determined by neutron activation analysis (NAA). The analysis method irradiates 0.2–0.4 g samples at ORNL's High Flux Isotope Reactor for approximately 10 seconds. The analysis itself is reported with an error that is determined using the uncertainties associated with the radiochemical analysis technique. Sampling of I–AgZ beds as performed here has been observed to contribute to uncertainty in the iodine content of the segment. The variability of a single 0.2 g subsample from the larger 5–8 g sample 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. 2017). The error associated with NAA is presented in the figures of this report, but the effects of sample heterogeneity should be also be considered during data analysis.

2.1 Thin-bed Iodine Loading of Aged AgZ

A sample of the AgZ recovered from the 8-month VOG test in 2018 was tested for sorbent capacity. The sample was recovered near the effluent side of the 2018 sorbent bed, and NAA confirmed that no iodine was adsorbed by AgZ near this edge during testing. The sorbent capacity of this 8-month aged sample was tested in a TGA by exposing a thin bed (1.7 g) of the sample to a dry air stream containing ~50 ppm iodine at 150°C until the loading curve flattened. The loading test of the 8-month aged sorbent took <80 hours to reach sorbent saturation.

2.2 Determination of Balance Gas Effects on AgZ Aging

Two 28-day tests were completed at ORNL. Both tests exposed a deep AgZ bed to a humid carrier gas stream with 1200 ppb CH_3I: the first test used nitrogen as the diluent gas, and the second used purified air. Both sorbent beds contained approximately 62 g of AgZ, which was sectioned into 10 segments separated by glass wool (Figure 1). Each segment contained between 3 and 9 g of sorbent, and the segment mass generally increased from the base to the top of the bed. The sorbent beds were held at 150°C. The inlet air stream was humidified by sparging a bubbler of water with either air or nitrogen at a set flow rate that resulted in an inlet dew point between 8 and 9°C.

A CH_3I stream was generated using a Kin-Tek Flexstream Gas Generator containing five permeation tubes with emission rates of ~6000 ng/min each. Calculations indicated that 1.2 g of iodine (as methyl iodide [CH_3I]) should have been released from the permeation tubes during the test period. Gravimetric determination of mass loss during the test confirmed that 1.3 g of CH_3I were released from the tubes and delivered to the beds.

After the test was completed, the sorbent was removed from the column by vacuum, and each section was gently mixed before sampling for NAA. The segments are labeled alphabetically from A (base) to J (top) (Figure 2; segments F through J are hidden by the column clamp).

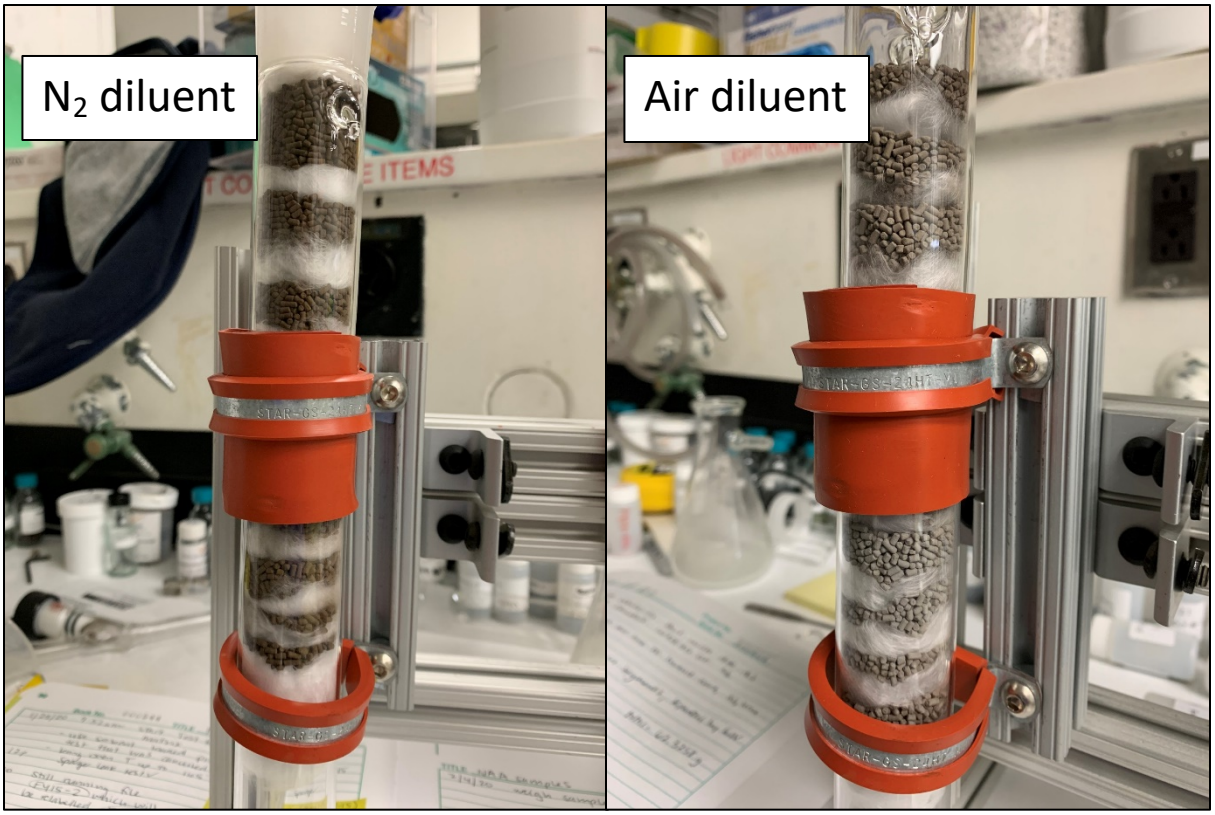

Figure 1. Sorbent beds used in the aging tests, shown after the completion of the test. The nitrogen diluent test is shown on the left and the air diluent test is shown on the right.

2.3 Extended-duration VOG Testing

Two extended VOG tests were completed in parallel at ORNL between May 30, 2019, and March 3, 2020, for a total of 279 days or 9 months. Approximately 139 g of sorbent was loaded into each column for a total bed depth of ~9.5 inches (Figure 2). The first test contained 50 ppb I_2 in a humid air stream, and and the second contained 150 ppb CH_3I in a humid air stream. The sorbent beds were held at 150°C. The air was humidified by sparging dry air (−70°C dew point) at a known rate through a water bubbler. The I_2 and CH_3I streams were generated using Kin-Tek Flexstream Gas Generators. One I_2 permeation tube was used with an emission rate of 2065 ng/min at 100°C, and two CH_3I permeation tubes were used with a total emission rate of 3179 ng/min at 30°C. Periodically, all flows would be suspended for <3 min while the bubbler was refilled with water.

The sorbent beds were not segmented during initial loading, so segments were created at the end of the test by vacuuming out sections of the bed from the top-down. Each section was gently mixed before sampling for NAA. The segments are labeled alphabetically from A0 (base) to J (top).

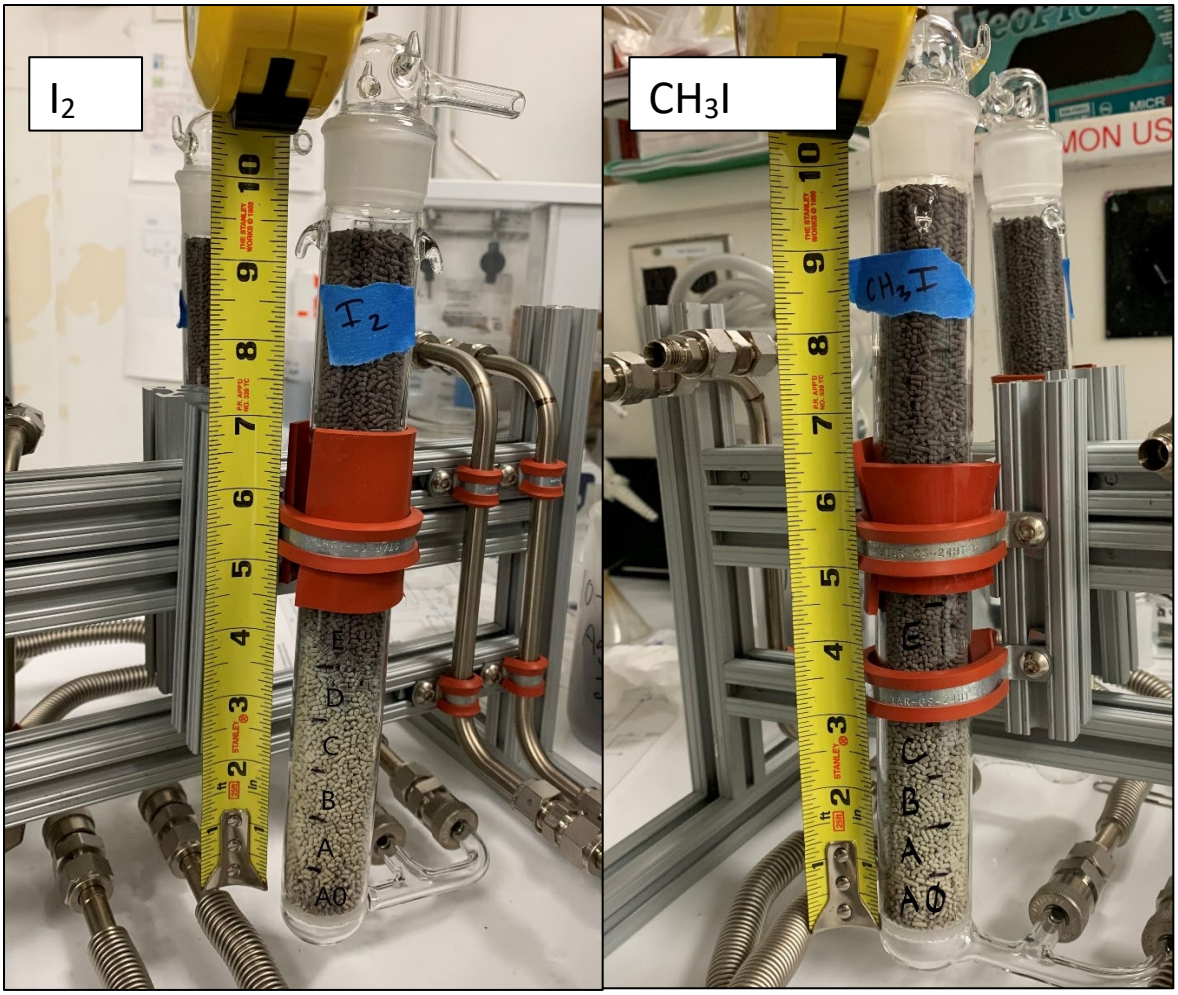

Figure 2. The I_2 and CH_3I sorbent beds at the end of 9-month VOG testing. The lower segments are labeled from A0 (base) to E (middle of column).

3. RESULTS

Three sets of experiments were performed to determine how sorbent capacity varies from aging under different timescales and gas compositions. The first tested the capacity of sorbent aged for 8 months under a humid air stream. The second tested differences in sorbent capacity and MTZ length during high-concentration CH_3I loading in a humid N_2 stream and a humid air gas stream over 28 days. The third tested sorbent capacity and MTZ length during low concentration CH_3I and I_2 loading in a humid air stream over 9 months.

3.1 Capacity of 8-month Humid Air-Aged AgZ

The iodine capacity of the 8-month humid air-aged AgZ was determined to be 35 mg I/g AgZ using thin-bed testing. The iodine loading determined by TGA is shown in Figure 3. Some discontinuity is observed in this loading curve, perhaps owing to the unstable nature of the balance under changing room temperatures, but the final iodine loading was confirmed through NAA.

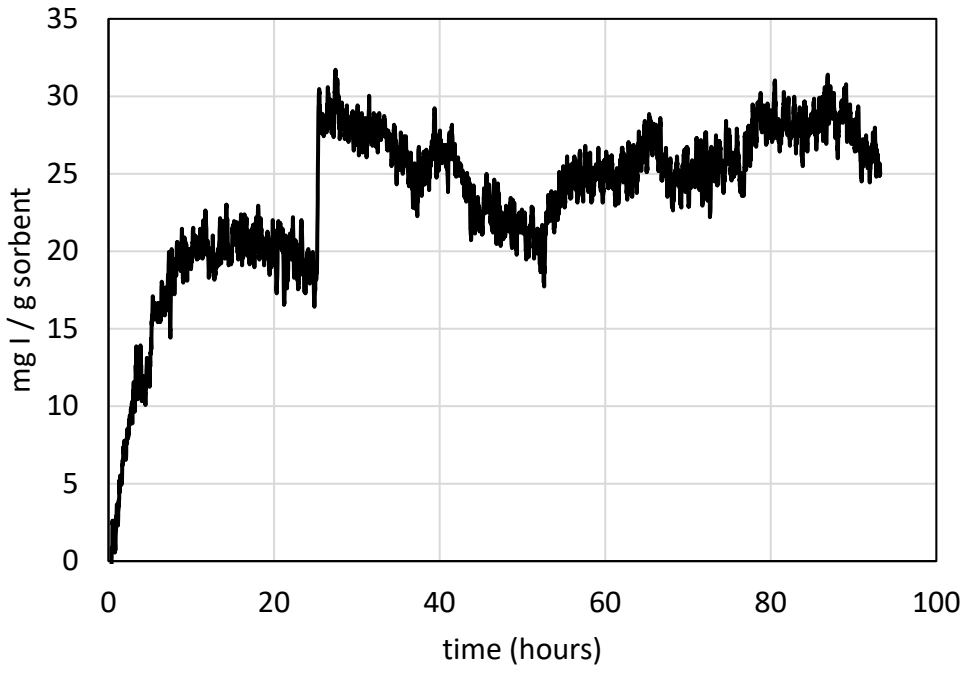

Figure 3. Iodine (I_2) adsorption by 8-month humid-aged AgZ.

3.2 Balance Gas Effects

The appearance of the two sorbent beds was noticeably different at the conclusion of the test (Figure 1). The N_2-diluent test resulted in almost no sorbent color change, but the air-diluent test resulted in a color change from brown to light gray-green.

Iodine was measurable in all bed segment samples from both tests. The minimum detectable amount of iodine reported by NAA was between 0.001 and 0.002 wt% I, or 0.01 and 0.02 mg I/g AgZ. The mass of each segment was used to calculate the volume and height of the segment using the average AgZ bulk density of 0.866 g/cm^2. These data are presented in Table 1.

Table 1. Iodine loading of AgZ sorbent beds as a function of bed depth and balance gas.

Diluent gas	Segment	Segment length (cm)	Height from base (cm)	Iodine loading (mg I/g AgZ)	+/-
N_2	A	0.7	0.7	46.1	5.00
N_2	B	0.8	1.5	47.7	5.15
N_2	C	1.2	2.7	47.1	2.20
N_2	D	1.2	3.9	39.8	4.34
N_2	E	1.3	5.2	33.4	3.66
N_2	F	1.8	7.0	26.8	2.95
N_2	G	1.9	8.9	20.9	2.32
N_2	H	2.0	10.9	16.7	1.85
N_2	I	1.8	12.7	10.1	1.14
N_2	J	1.2	13.9	7.46	0.84
air	A	0.7	0.7	45.6	0.98
air	B	0.7	1.4	45.6	0.93
air	C	1.2	2.6	41.1	0.85
air	D	1.5	4.1	32.3	0.68
air	E	1.6	5.6	24.3	0.52
air	F	1.9	7.6	15.7	0.34
air	G	1.9	9.5	10.7	0.24
air	H	1.9	11.3	7.44	0.17
air	I	1.3	12.6	3.53	0.09
air	J	1.3	13.9	1.47	0.04

The maximum loading observed in the nitrogen test is 47.7 mg I/g AgZ (segment B) and the minimum is 7.5 mg I/g AgZ (segment J). The iodine loading of the first four bed segments from the N_2 test are within the margin of error of each other, so the bed is considered saturated on the leading edge (Figure 4). The maximum loading observed in the air test is 45.6 mg I/g AgZ in the first two segments, and the minimum loading is 1.47 mg I/g AgZ in the top segment (J). Sorbent saturation of the leading edge was also observed in the air test, with the first three segments reporting the same iodine loading within error margins. In Figure 4, discrete bars indicate segment length. The uncertainties in iodine loading are reflective of the error associated with NAA and do not include any uncertainties derived from sampling.

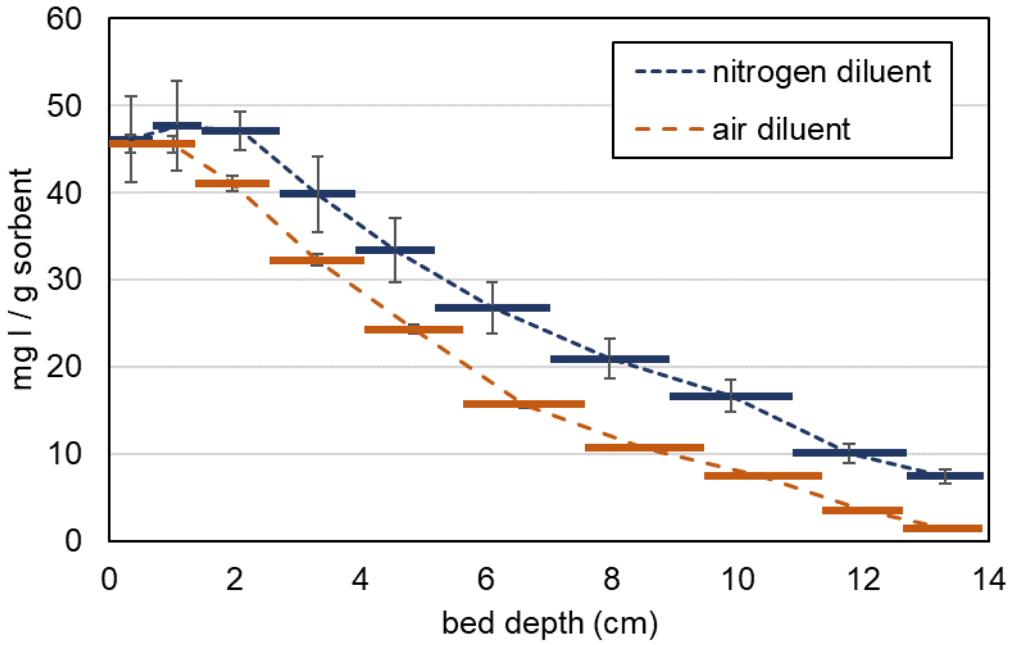

Figure 4. Iodine loading during 28-day aging tests using a nitrogen diluent and air diluent.

No significant differences were observed between the two tests, indicating that the use of nitrogen as a balance gas does not mitigate the aging of AgZ. Visually, the shape of the adsorption front within the bed is steeper for air. However, because the heterogeneous nature of iodine loading on the sorbent and the lack of direct CH_3I concentration measurements at the bed inlet, the results of the two tests are considered to be quite similar.

3.3 Extended-duration VOG Tests

The sorbent columns after 9 months of aging are shown in Figure 2. Sorbent discoloration was observed over a longer column length for the CH_3I test compared to the I_2 test. When dismantling and segmenting the sorbent columns, channeling along the outer edge of the column was inferred because the sorbent in contact with the glass was more discolored (light green) than the sorbent near the middle of the column (Figure 2). The flow path of the air stream appears to have resulted in the iodine depositing on the sorbent in contact with the glass first, with loading moving inward toward saturation. The segment at the base of both columns (A0) showed heterogeneous discoloration, but the two segments above the base (A and B) were mostly a homogeneous light green color.

Loading data from the extended VOG tests are presented in Table 2. Iodine was measurable in the lower six bed segments from both the CH_3I test and I_2 test. Above these segments (12.4 cm for CH_3I and 12.8 cm for I_2), iodine loading was less than 0.02 mg I/g AgZ. Neither deep bed appears to have reached sorbent saturation with the maximum iodine concentration observed in the bottom segment of both tests with no flattening of the loading curve at the base of the bed (Figure 6). The maximum loading observed in the CH_3I test is 57.4 mg I/g AgZ, and the maximum loading observed in the I_2 test is 46.8 mg I/g AgZ.

Table 2. Iodine loading of AgZ sorbent beds following extended VOG tests.

Iodine species	Segment	Segment length (cm)	Height from base (cm)	Iodine loading (mg I/g AgZ)	+/-
CH_3I	A0	2.4	2.4	57.35	1.21
CH_3I	A	2.0	4.3	34.70	0.74
CH_3I	B	2.3	6.6	18.27	0.39
CH_3I	C	2.1	8.7	4.33	0.11
CH_3I	D	1.9	10.6	0.82	0.02
CH_3I	E	2.3	12.8	0.12	0.01
CH_3I	F	4.3	17.1	0.01	<0.01
CH_3I	G	2.6	19.8	0.01	<0.01
CH_3I	H	2.0	21.8	0.01	<0.01
CH_3I	I	1.9	23.7	0.02	0.02
CH_3I	J	2.4	26.1	0.01	<0.01
I_2	A0	2.0	2.0	46.85	0.94
I_2	A	2.5	4.5	26.00	0.54
I_2	B	2.0	6.5	14.64	0.31
I_2	C	2.3	8.8	5.16	0.12
I_2	D	1.9	10.7	1.82	0.04
I_2	E	1.7	12.4	0.06	0.01
I_2	F	2.3	14.6	0.01	0.01
I_2	G	2.2	16.8	<0.01	<0.01
I_2	H	3.4	20.3	0.01	<0.01
I_2	I	2.5	22.8	0.01	0.01
I_2	J	3.0	25.8	0.01	<0.01

4. DISCUSSION

4.1 Effects of Aging on Sorbent Capacity

The measured iodine loading capacity for 8-month humid air-aged AgZ was synthesized with previously obtained results (Table 3) to generate the curve shown in Figure 5. The uncertainty associated with capacity measurements is assumed to be 10%, as described in Section 2.

Table 3. Capacity of AgZ as a function of humid air aging duration.

Aging Time (months)	Capacity (mg I/g AgZ)
0	108
1	50
2	42
4	40
8	35

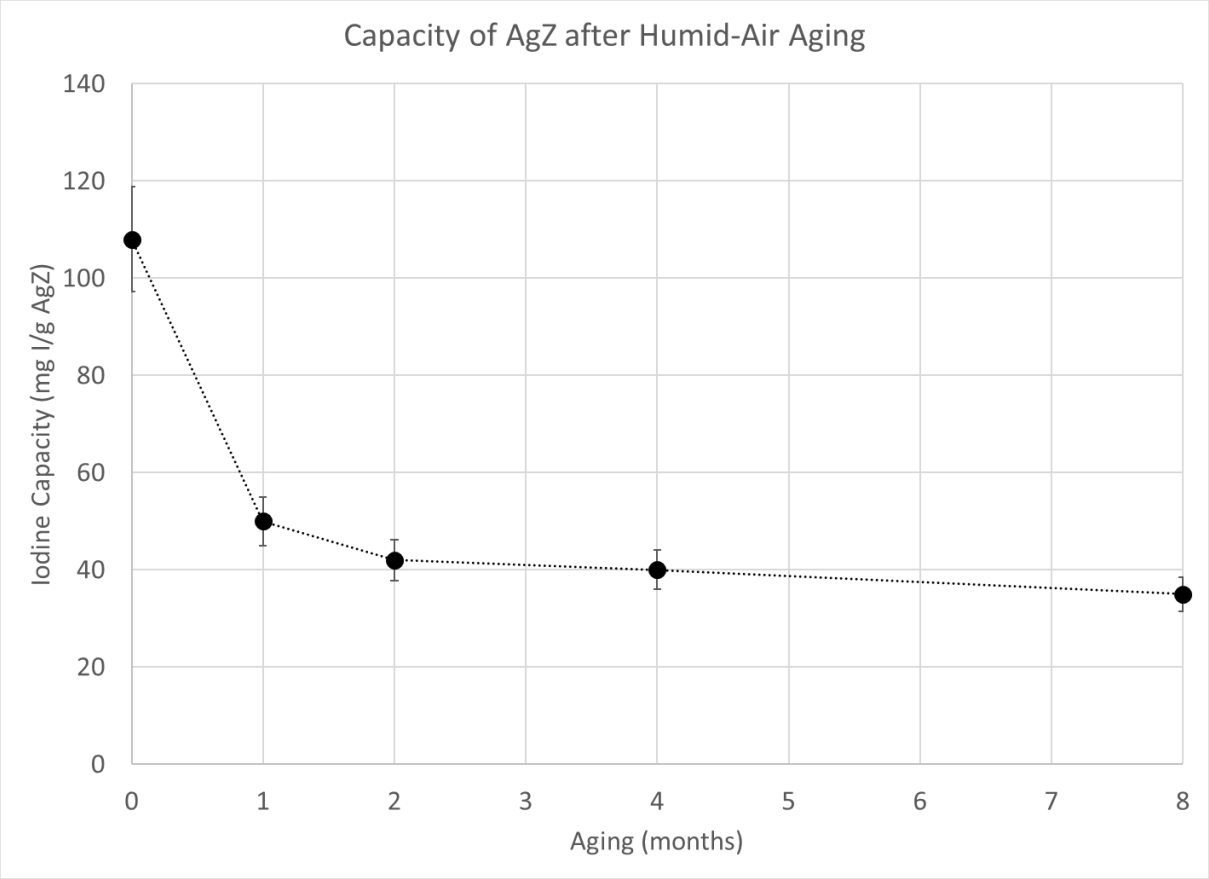

Figure 5. Iodine capacity of AgZ as a function of humid air exposure duration.

The bulk of measured capacity loss occurs during the first month of aging, with only marginal additional losses after 2 months of aging. Validating previously obtained aging data, the 28-day tests described in Section 3.2 showed iodine loadings similar to corresponding test results found in Table 3.

Aging of AgZ for 8 months in a humid air stream represents the longest test of this type and corresponds to the potential service duration for VOG beds (Jubin et al. 2016). Importantly, this test provides a lower bound on the capacity of aged AgZ. Although the conceptual VOG system designed by Jubin et al. (2016) assumed a maximum halogen loading of 1 wt%, the total halogen loading likely could be assumed to be closer to 2–3 wt% and still provide a reasonable engineering margin.

4.2 MTZ Estimations for Short-duration Testing

The nitrogen-diluent test achieved sorbent saturation (47 mg I/g AgZ) across the first 2.7 cm of sorbent, and the air-diluent test achieved sorbent saturation across the bottom (46 mg I/g AgZ) first 1.4 cm of sorbent, allowing estimation of the MTZ for these experimental conditions. After saturation, the slope of the MTZ was assumed to be linear through each bed. Using a linear regression of Segments D–J for the N_2 test and Segments C–J for the air test, the lengths of the MTZs were estimated at 16.0 and 13.4 cm, respectively. The linear regressions and R^2 values for each curve are found in Table 4.

Table 4. Estimation of MTZ for CH_3I adsorption by AgZ during 28-day aging tests.

Test	Segments used in regression	Linear equation	R^2	y-intercept MTZ length (cm)
N_2	C–J	$y = -3.14x + 50.17$	0.9896	16.0
Air	D–J	$y = -3.40x + 45.63$	0.9592	13.4

Test conditions: CH_3I concentration in feed stream = 1200 ppm. Inlet dew point of feed stream = 0°C. Sorbent bed held at 150°C. Superficial gas velocity through the bed was 10 m/min.

These are estimations but can provide a helpful first iteration for design purposes. An examination of Figure 4 shows that the air-aged sorbent appears to have some tailing on the leading edge of the bed. The slope of the mass transfer zone from the point of sorbent capacity to the top edge of the bed is similar between the two diluent streams: –3.1 (mg I/g AgZ)/cm for the nitrogen stream and –3.4 (mg I/g AgZ)/cm for the air stream.

4.3 Observations and MTZ Estimations for Extended-duration Testing

The maximum iodine loading observed for the extended-duration CH_3I test was 57.4 (±1.2) mg I/g AgZ; the maximum iodine loading observed for the extended-duration I_2 test was 46.8 (±0.9) mg I/g AgZ. There are no indications that either test resulted in sorbent saturation of the leading edge of the bed.

These data can be compared to previous VOG experiments. Jubin et al. (2018) completed an 8-month deep bed test characterizing iodine adsorption by AgZ at an inlet iodine concentration of 50 ppb_v I_2 Figure 6 shows the loading profile for the 2018 test along with the loading profiles observed during the testing described in this report. The 2018 test did not achieve sorbent saturation, with the highest recorded iodine concentration being 38.8 (±0.6) mg I/g AgZ at the base of the bed. Given the maximum concentration of 46.8 mg I/g AgZ recorded in the 9 month test presented here, it can be inferred that after 8 months online, the rate of iodine uptake by AgZ is ~6 mg I/g AgZ per month.

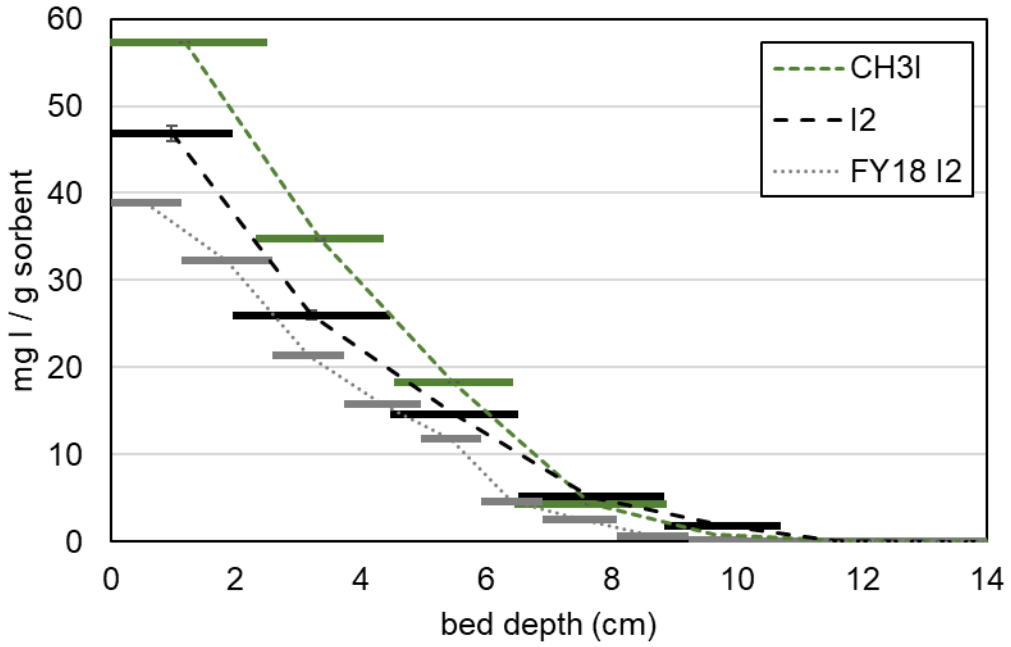

Figure 6. Iodine (black) and CH_3I (green) loading during 9-month extended VOG tests. Iodine loading from an 8-month VOG test is compared (grey, Jubin et al. 2018).

Modeling exercises by Jubin et al. (2018) assumed that aged-AgZ capacity for I_2 loading is 45 mg I/g AgZ, which is very close to the iodine loading levels observed in this testing and is commensurate with results of the aging studies presented in Sections 3.1 and 3.2. If this assumption is applied to the extended-duration testing completed here, then it is likely that the I_2 bed was at or near sorbent capacity at the leading edge of the bed. If it is further assumed that the CH_3I bed was at or near sorbent capacity at the leading edge of the bed, then the MTZ can be assessed for each iodine species as the length from the leading edge of the sorbent bed to the point where iodine loading is below NAA detection limits. This results in the MTZ for CH_3I adsorption by AgZ under these test conditions being estimated as 12.8 cm, and the MTZ for I_2 adsorption being estimated as 12.4 cm (Figure 6). However, these lengths are considered preliminary until testing can confirm sorbent saturation levels and better reveal the geometry of the loading curve.

5. CONCLUSIONS

A comparison of the aging tests presented here and those from Jubin et al. (2012a) show that the capacity of AgZ drops to ~50% after 1 month of humid aging, and further drops to ~40% capacity after 4 months of humid aging. The capacity does not decrease further up to 8 months of humid aging. The drop in capacity after 1 month of humid aging is similar for CH_3I loading (~45% decrease) and I_2 loading (~55% decrease; Jubin et al. 2012a). One-month aging tests that compared nitrogen and air as the gas stream diluent resulted in similar maximum loading capacities and overall loading curves, indicating that the effects of aging cannot be mitigated by avoiding air as the balance gas.

Nine-month extended duration testing of I_2 (50 ppb_v) and CH_3I (150 ppb_v) adsorption by AgZ under VOG conditions resulted in maximum sorbent loading of 46.8 and 57.4 mg I/g AgZ, respectively. Saturation of the leading edge of each bed was not observed, but the aging data described above would indicate that the sorbents were near saturation. Using this assumption, the MTZ for iodine adsorption by AgZ at parts per billion concentrations was estimated to be 12.4 cm for I_2 and 12.8 cm for CH_3I. The slope of the iodine

loading curve and the length of the estimated MTZ correlate well with a similar 8-month VOG test completed in 2018 (Jubin et al. 2018).

The MTZs estimated for 1-month humid-aged AgZ were 13.4 and 16.0 cm with an inlet CH_3I concentration of 1200 ppb_v, somewhat longer than was estimated for the extended duration testing that contained 150 ppb_v CH_3I in the feed stream. This may indicate that iodine concentration of the feed stream has notable effects on the length of the MTZ. This possibility is supported by deep bed testing conducted at Idaho National Laboratory in which CH_3I concentration of the feed stream was observed to have a significant effect on the length of the MTZ at parts per million concentration (Bruffey et al. 2019).

The data collected here do not support the use of N_2 balance gas as a way to mitigate aging effects and conduct accelerated VOG testing. It does provide updated information on the total iodine capacity of AgZ after humid-aging for durations relevant to full-scale UNF reprocessing. It also increases the available information regarding the length of the mass transfer zone. It is recommended that future work re-evaluate the design of the VOG iodine capture system provided by Jubin et al. (2016) with this updated data. It is expected that the total sorbent consumption will be decreased and that the bed changeout frequency may be increased, decreasing the cost of the iodine abatement system needed for mitigation of iodine within the VOG stream.

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