Data Requirements and Test System Needs for Development of an Integrated Off-Gas Treatment System

Nuclear Technology Research and Development

Prepared for
U.S. Department of Energy
Material Recovery and Waste Form Development Campaign
R. T. Jubin, S. H. Bruffey, J. A. Jordan, B. B. Spencer
Oak Ridge National Laboratory
N. R. Soelberg, A. K. Welty and M. Greenhalgh (INL)
6/30/2017
NTRD-MRWFD-2017-000422
ORNL/TM-2017/314

Approved for public release. Distribution is unlimited.

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise, does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

SUMMARY

In fiscal year 2016 an Engineering Evaluation of an integrated off-gas system was conducted. This study resulted in a report entitled Engineering Evaluation of an Integrated Off-Gas Treatment System for Used Nuclear Fuel Reprocessing Facilities (Jubin et al. 2016a), from here referred to as the "Engineering Evaluation." This study focused on the capture and retention of the volatile radionuclides (^{3}H, ^{14}C, ^{85}Kr, and ^{129}I), selected semi-volatile radionuclides, specifically ^{106}Ru, and chemical species like NO and NO_2 generated during the dissolution of the used nuclear fuel. The study examined the design of the combined head-end off-gas streams and the vessel off-gas stream. The study drew upon the available literature to conduct the equipment sizing and sorbent usage. A number of assumptions were required to complete this analysis and major gaps in the available data were identified that, if resolved, could increase the fidelity of the engineering design.

This assessment looks at those identified data gaps and the more subtle assumptions that were required in the Engineering Evaluation and provides a detailed look at the specific data needs for each major system. This assessment of the data gaps builds on a study conducted early in fiscal year 2016 that established a set of performance criteria for capture and immobilization technologies. The gaps identified in the engineering assessment also provided a check on the breadth of the criteria and metrics that were previously developed. During the analysis of the data gaps it was necessary to expand the metrics slightly to capture co-adsorption effects and to address desorption, neither of which were included in the original set of metrics.

Across the six major off-gas control systems evaluated a number of common data gaps became apparent. These include:

- Capacity and adsorption rate data needs appear common for virtually all the unit operations considered. These two parameters dictate the dimensions and operating conditions of the sorbent beds and wet scrubber systems. The capacity dictates the mass and volume of Ru, tritium, I, and Kr/Xe sorbent beds for a given adsorption duration; the adsorption rate dictates the needed depth of the solid sorbent beds and the height of the wet scrubbers. Co-adsorption of other species is important to the extent that this interferes with the adsorption of the target species, impacts process operation, or impacts handling, recycle, or disposal of the spent sorbent/scrub solutions, and the waste form and disposal of the captured species. This is of secondary importance compared to demonstrating and optimizing capacity and adsorption rates for the target species; but co-adsorption and impacts of co-adsorption are areas where generally even less is currently known for both sorbent beds and wet scrubber systems.
- Sorbent particle and bulk densities impact the volume of the adsorption system for a given mass of sorbent, but these physical properties data are readily measured.
- For systems including regenerable sorbents, data on desorption and multicycle operations is also critical for the sizing of the subsequent recovery systems, the design of any additional purification steps, and the conversion to a waste form. In general, obtaining this type of data requires larger, more complex test beds and must be able to test the effects of co-adsorbed species. Regenerable sorbents are being tested for the separation of Kr and Xe and the removal of tritium. The scrub solution used in the CO_2 scrubber is recycled back to the scrubber after precipitating out the captured CO_2 and may also be considered regenerable.

Many of the data gaps identified in this report can be effectively addressed with test systems focused on specific target components. In the case of the regenerable sorbents that focus on tritium, Xe and Kr, more complex test beds will be required to obtain data on the desorption behavior, co-absorbed species effects, potential product purification, and cycle time. In the case of Xe and Kr some preconditioning of the gas streams may also be required in an actual plant design.

This page is intentionally left blank.

CONTENTS

This page is intentionally left blank.

FIGURES

TABLES

This page is intentionally left blank.

ACRONYMS

AgZ	silver-exchanged mordenite (zeolite)
AgZ-PAN	silver-exchanged mordenite (zeolite)-polyacrylonitrile
CETE	Coupled End-to-End Research Project
DF	decontamination factor
DOG	dissolver off-gas
FY	fiscal year
HEPA	high-efficiency particulate air
HZ-PAN	hydrogen mordenite (zeolite)-polyacrylonitrile
MOF	metal organic framework
MS	molecular sieve
MTZ	mass transfer zone
ORNL	Oak Ridge National Laboratory
R&D	research and development
TGA	thermal gravimetric analyzer
TPT	tritium pretreatment
TPTOG	tritium pretreatment off-gas
VOG	vessel off-gas

This page is intentionally left blank.

DATA REQUIREMENTS AND TEST SYSTEM NEEDS FOR DEVELOPMENT OF AN INTEGRATED OFF-GAS TREATMENT SYSTEM

1. INTRODUCTION

In fiscal year (FY) 2016 an Engineering Evaluation of an integrated off-gas system was conducted. This study resulted in a report entitled Engineering Evaluation of an Integrated Off-Gas Treatment System for Used Nuclear Fuel Reprocessing Facilities (Jubin et al. 2016a), from here referred to as the “Engineering Evaluation.” This study focused on the capture and retention of the volatile radionuclides (^{3}H, ^{14}C, ^{85}Kr, and ^{129}I), selected semi-volatile radionuclides, specifically ^{106}Ru, and chemical species like NO and NO_2 generated during the dissolution of the used nuclear fuel. The study examined the design of the combined head-end off-gas streams and the vessel off-gas (VOG) stream. The study considered two primary cases: (1) the direct dissolution of the fuel without tritium pretreatment (TPT) and (2) the use of air TPT. In the case of direct used nuclear fuel dissolution, the shear off-gas is routed to a dissolver and the resulting dissolver off-gas (DOG) stream is treated as shown in Figure 1. In the case of the air TPT, the off-gas from TPT is treated separately to recover the tritium and Ru before joining the off-gas from the dissolver between the dual condensers and the iodine recovery beds as shown in Figure 2. The VOG system is shown in Figure 3. These three figures are shown as they were originally presented in Jubin et al. 2016a and include stream numbers that will not be further discussed in this report.

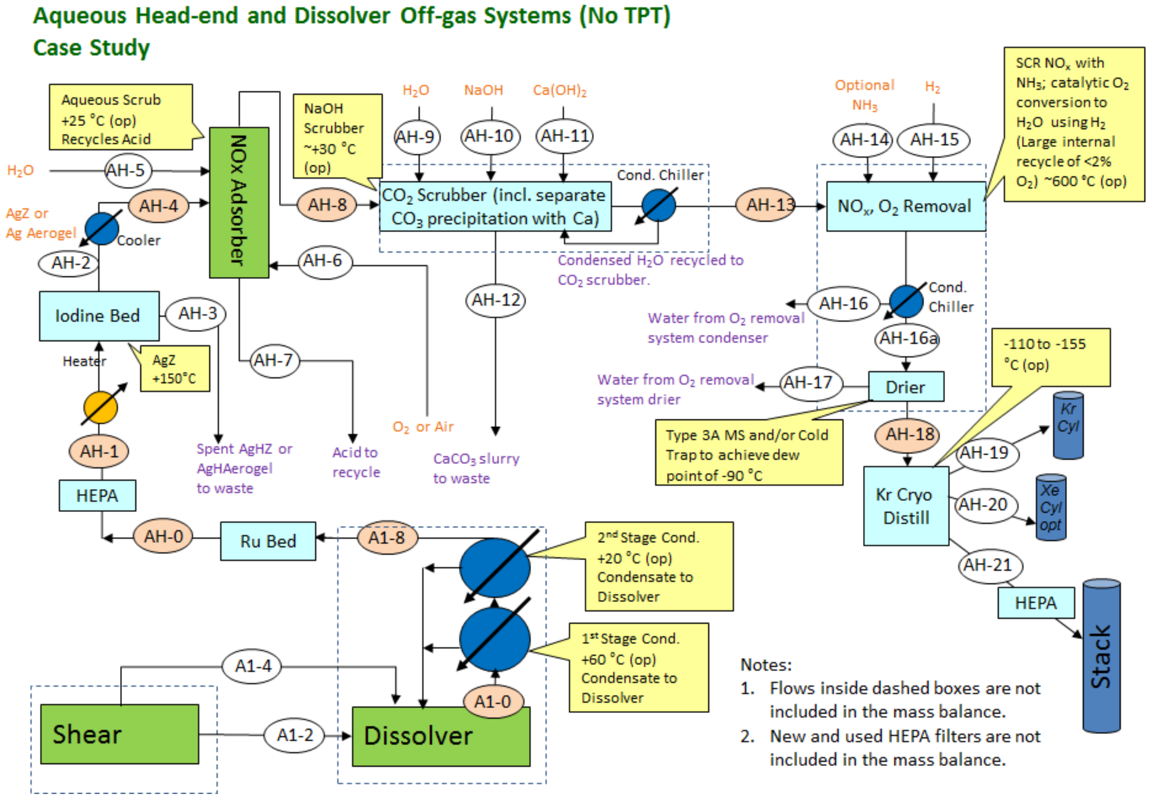

Figure 1. Block diagram for a dissolver off-gas treatment system (Jubin et al. 2016a).

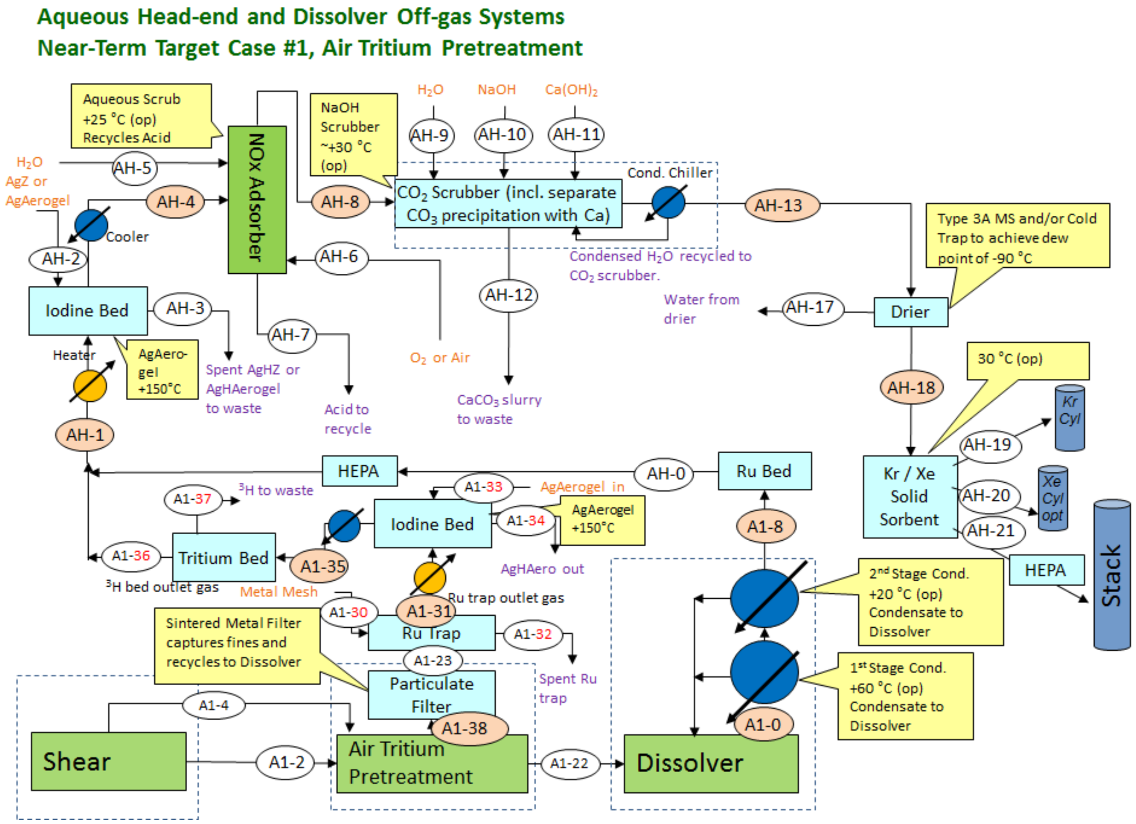

Figure 2. Block diagram for a dissolver off-gas and TPT off-gas treatment system (Jubin et al. 2016a).

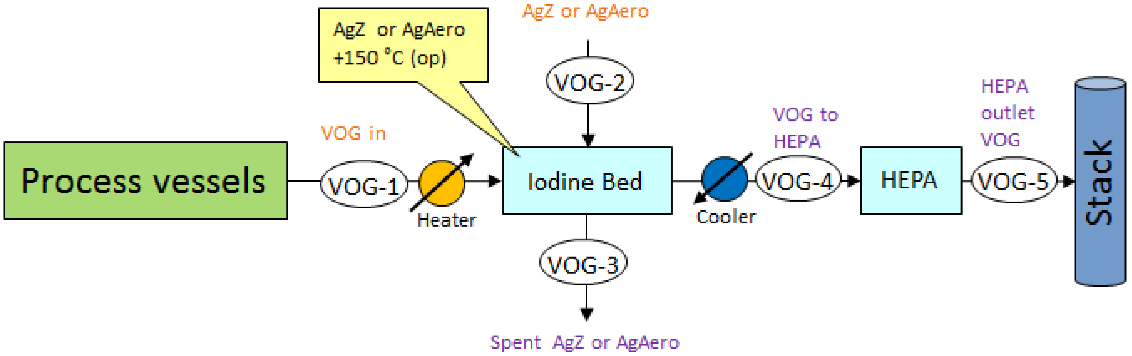

Figure 3. Block diagram for a vessel off-gas treatment system (Jubin et al. 2016a).

The study drew on the available literature to conduct the equipment sizing and sorbent usage. A number of assumptions were required to complete this analysis, and major gaps in the available data were identified that could increase the fidelity of the engineering design if resolved. This assessment looks at those identified data gaps and the more subtle assumptions required in the Engineering Evaluation, and attempts to provide a detailed overview of the specific data needs for each major system. This assessment provides a consolidated list of data needs, testing approaches, and equipment needs to close the gaps, as well as prioritizes the gaps to be closed.

2. APPROACH TO ASSESSING DATA GAPS

2.1 Framework for the assessment

This assessment of the data gaps will build on a study conducted early in FY 2016 that established a set of performance criteria for capture and immobilization technologies (Jubin et al. 2016b). Five criteria were identified for the capture material and related systems. These were (1) technical performance and characteristics (physical and chemical properties), (2) technical practicality, (3) system design and performance, (4) technical maturity, and (5) cost. For each of these five criteria, multiple metrics were identified to describe the important aspects of that criterion. Table 1 lists the five criteria and the associated metrics. Although these criteria were developed in part to provide a technically based foundation for the comparison of sorbents, it is believed that these criteria also could be used to provide a framework for the collection and assessment of the data gaps identified in the engineering assessment. The gaps identified in the engineering assessment should also provide a means to evaluate the breadth of the previously developed criteria and metrics.

2.2 Scope of data gap assessment

This assessment only examines the gaps associated with the design of the off-gas treatment equipment. It is fully recognized that there are also significant gaps in relevant source terms for both the dissolver and TPT systems. These gaps should be addressed by the research and development (R&D) efforts that are focused on the relevant processes.

3. IDENTIFIED DATA GAPS

During the Engineering Evaluation, a number of significant data gaps in the available information were identified. The gaps noted in the Engineering Evaluation are listed below with no importance assigned to the order (Jubin et al. 2016a):

- Pressure swing regeneration of the xenon and krypton columns—This technique appears to be viable, possibly more viable than temperature swing because of the short column change-out times required. Investigation of this technique is lacking in the current research plan.
- Face velocity values—There has been little research into what face velocities can be tolerated by the sorbents, particularly for the metal organic framework (MOF) materials.
- Mass transfer zone (MTZ) determinations—Very limited data exist on the length of the MTZ for silver-exchanged mordenite (AgZ) or Ag-Aerogel. MTZ lengths for both AgZ and Ag-Aerogel have been estimated from deep-bed tests for several years, but these estimates are limited to test durations of up to several hundred hours operation. The variation in gas flow rates, temperatures, and compositions has been limited to the scopes of those tests such as has been reported in Soelberg et al. 2013; Soelberg and Watson 2014, 2015, and 2016; and Soelberg 2016. There are few to no data on how this is impacted by gas rate and concentration. Similar measurements are lacking for the MOF materials. Testing of PAN materials has shown little velocity effect up to 6.4 m/min, but higher velocities have not been tested (Welty et al. 2016).
- Ruthenium decontamination factor (DF) values—An overall ruthenium DF value of 3.4×10^7 is needed for 5 y cooled 60 GWd/MTIHM fuel. A review of the literature supports a DF value of about 1×10^4. Some uncertainties present in the literature lead to this difference. The main uncertainty is the conversion of volatile RuO_4 to particulate RuO_2 in the gas phase and if volatile and particulate forms of ruthenium can be reliably captured and with what efficiency. This applies to various off-gas processes, including filters, adsorbers, condensers, and wet scrubbers, that are not specifically

intended for ruthenium capture. The efficiencies of high-efficiency particulate air (HEPA) filters for particulate RuO_2 and of aqueous scrubbers for volatile RuO_4 are uncertain. A critical review of the literature and proposed research to close the gaps identified in this review are needed.

- Volatilization of ruthenium—Improved data are needed on the volatilization of ruthenium during TPT. Data about the relationship of quantity released, speciation, and operation conditions (temperature, time, oxygen content etc.) are needed.
- Capacity of metal mesh for Ru—No data exist on the capacity of metal mesh for the capture of ruthenium; thus, sizing of this alternate approach could not be completed.
- Desorption of physisorbed species—Physisorbed iodine exists on iodine-loaded AgZ and, to a lesser extent, on Ag-Aerogel. The quantities are uncertain but important to the operation of the removal process.
- Verification of MOF operating processes—The operating scheme detailed for the MOF, hydrogen mordenite-polyacrylonitrile (HZ-PAN), and silver-exchanged mordenite-polyacrylonitrile (AgZ-PAN) sorbents has not been fully tested and should be verified experimentally.
- Optimize krypton and xenon desorption—Desorption of krypton and xenon from PAN and MOF has not been optimized in terms of product purity, segregation from co-adsorbed gases, and energy balance. In particular, for the solid xenon and krypton sorbents, co-adsorption of air and other off-gas components impacts the purity of the target gases that are released or stored. Few data are available on this topic, but low sorption of these air and off-gas components on the sorbent has the potential to greatly simplify the removal of xenon and krypton from the off-gas stream, especially if the additional krypton product purification process can be eliminated or simplified.

Table 1. Capture media criteria and metrics (Jubin, et al. 2016b)

Property	Unit	Desired trend	Comments
Metrics for technical performance and physical and chemical characteristics criterion			
Capacity	mol/m^3	High	The capacity of the bulk material for the radionuclide of interest. Capacity and bulk density influence sorbent column size.
Selectivity	$(X_a/Y_a)/(X_b/Y_b)$ (unitless) *Where* X_a *and* X_b *are mol fractions of species a and b respectively in the adsorbed phase, and* Y_a *and* Y_b *are mol fractions of species a and b in the bulk phase.*	High	The extent to which the target element is concentrated in the capture system compared with non-targeted elements. It influences how much preprocessing of the incoming off-gas stream must be done to make the material practicable. It also dictates how much post-processing may be required to separate isotopes that should not be mixed (e.g., ^{3}H and ^{129}I) before conversion to a final waste form.
Particle density	kg/m^3	High	Particle density is the density of the sorbent media and included internal pore volume. This is in contrast to the bulk density that is the average density of the sorbent bed taking into account the inter-particle void volume. Density, along with capacity, influences sorbent column size. There is a balance between the density and the permeability of the sorbent bed that should be considered and may set an upper density limit. High densities can negatively affect the sorption kinetics of the target element on the sorbent.
Surface area	m^2/g	High	Increased surface area can promote the efficiency of the sorbent. This is not the geometric surface area of the particles, but the gas active surface area, as likely measured by the Brunauer–Emmett–Teller method.
Specific heat capacity	J/(K·kg) or J/(K·m^3)	Application dependent	In use, as sorbates load onto the sorbent, the heat of reaction can cause the temperature of the sorbent to rise. Heat capacity can mitigate this rise. It also affects the time required to preheat or cool a sorbent column and so may have implications for process design.
Thermal conductivity	W/(m·K)	High	The thermal conductivity should be sufficiently high that the heat of reaction or decay can be dissipated sufficiently to maintain a desired bed temperature and to avoid significant thermal gradients within the sorbent bed. This is a key property for the design and size of systems that need to be heated or cooled.
Radiation stability	% degradation in capacity over time as a function of radiation exposure	High stability; low degradation over time	Intense background radiation and radioactive sorbates can cause substantial damage to the sorbent, especially those isotopes with high specific activity (^{3}H and ^{85}Kr). This can affect basic sorbent properties (such as capacity, selectivity) and sorbent lifetime.

Table 1. Capture media criteria and metrics (Jubin, et al. 2016b) (continued)

Property	Unit	Desired trend	Comments
Mechanical stability	Generated fines <420 μm with losses to the off-gas stream of <50 μg/m^3	High stability; low fines generation	High gas velocities, chemical reactions, and other plant variables can cause the bed packing to vibrate and cause some attrition of the particles that make up the bed. Attrition can yield fine airborne particles that may affect downstream processes, cause premature failure of the HEPA filters, and, perhaps, affect facility DF. The ability to limit these losses is reflected by a particle's mechanical stability.
Thermal stability	% degradation in capacity over time at selected operating temperature	High stability; low degradation over time	Thermal stability over both the normal operating and process upset condition temperature ranges is necessary.
Chemical stability	% degradation in capacity over time as a function of other species present in gas stream	High stability; low degradation over time	Impacts the operating life and performance of the sorbent.
Reactivity	Compatibility as determined by standardized compatibility tables	Demonstrated compatibility with all components of gas stream and materials of construction	A measure of the interaction between the sorbent and materials of construction and other gas stream components.
Metrics for technical practicality criterion			
Regeneration	No. of cycles before degrading to 80% of capacity for the target element	High	This property affects the overall plant design and cost of operation. Some materials may not be regenerated, i.e., they are single use.
Bulk density	kg/m^3	High	Bulk density is the average density of the sorbent bed taking into account the particle density and the intra-particle void volume, i.e., the mass of the sorbent (kg) divided by the volume of the packed bed (m^3). Bulk density impacts bed size, pressure drop, and surface area available for reaction. Increases in bulk density approaching the particle density in the limiting case will result in the smallest bed size but excessive pressure drop.
Co-adsorbed species	mol/kg	Small in number of species and quantity	Indicates how much the overall capacity of the sorbent is affected by sorption of non-targeted elements. Tramp elements or isotopes (e.g., Cl, ^{12}C, Xe, and so on) fall into this category.

Table 1. Capture media criteria and metrics (Jubin, et al. 2016b) (continued)

Property	Unit	Desired trend	Comments
Robustness	% variation in operating parameters tolerated without deleterious effects	High	Tolerance to process upset conditions.
Flexibility and pretreatment	Operating ranges; no. of unit operations for pretreatment	High flexibility; minimal pretreatment	The width of the sorbent standard operating envelope.
Process complexity	No. and type of control systems and unit operations required.	Low	Affects the cost, volume, and footprint of the treatment system.
Energy consumption	kW/mol	Low	This is the energy required to effect separation of the target element.
Environmental safety and health	Classification according to National Fire Protection Association ratings	Low	Other applicable hazard classification systems may be used as appropriate (e.g., safety data sheet, American Society for Testing and Materials, Uniform Fire Code, International Fire Code, American National Standards Institute, and local and state codes).
Metrics for system design and performance criterion			
Pressure drop	Pa/m	Low	Pressure drop should be minimal to avoid operational difficulties. This value is affected by sorbent densities and bed size.
Decontamination factor (DF)	$[\text{Isotope}]_a/[\text{Isotope}]_b$ (unitless)	High	Regulatory requirements dictate the value for specific isotopes.
Bed volume	m^3	Low	Bed volume is a function of the capacity, particle density, and bulk density of the sorbent material.
Metrics for technical maturity criterion			
Technology readiness level of sorbent system	1–9	High	The technology readiness level is defined in the technology readiness assessment report (DoD 2011).
Commercial availability	Yes/No	Readily available	Commercial availability of the sorbent is an important parameter because of the cost of producing a material "in house." However, some sorbents can be made in place, such as a $AgNO_3$ solution on an inert substrate (e.g., Al_2O_3). Commercial availability eliminates the need to build infrastructure and accrues the benefit of scale because material is made for a number of customers.

Table 1. Capture media criteria and metrics (Jubin, et al. 2016b) (continued)

Property	Unit	Desired trend	Comments
Time to commercialization	y	Short	This must be compatible with the construction of the reprocessing facility.
Metrics for cost criterion			
Cost of sorbent material	\$/kg, \$/Ci, or \$/mol	Low	Cost of sorbent and any associated consumable materials.
Operating cost	\$/Ci	Low	Cost to operate the capture system.

It should be noted that as the data gaps were being enumerated it was necessary to expand the metrics slightly to capture co-adsorption effects and to address desorption. The metrics for bulk density and co-absorbed species were moved to the criterion that captured the metrics for chemical and physical properties. For the purposes of this evaluation, the criteria for technical maturation and cost were not considered.

4. DATA GAPS

4.1 Ruthenium recovery systems

During the preparation of the Engineering Evaluation it became apparent that very little data were available that could be directly applied to the design and sizing of a ruthenium recovery/abatement system. Limited data were available on the fraction of ruthenium volatilized either in the TPT system or dissolver, and speciation data was unavailable. Although these data are critical to the design of the recovery systems, these data should be obtained as part of the development of the TPT system and dissolver system, along with other relevant data that would further refine the off-gas source terms.

Table 2 summarizes the data gaps and the data requirements for the Ru capture system. These gaps follow the same structure as described in the criteria report but focus primarily on the properties of the sorbent materials and key operational parameters.

Because of the lack of data that can be used to design the systems, the most important data gap to be filled is to obtain data on the Ru capacity of the silica gel sorbent and selected metal mesh material as a function of Ru concentration in the off-gas stream (Gap ID #1). The next most critical is the determination of the co-absorbed species and their impact on the Ru adsorption capacity. (Gap ID #3 and #5). Of particular importance is the impact of water/acid vapor on the silica gel sorbent capture efficiency for systems similar to the dissolver off-gas. Addressing these three gaps will allow a significant refinement in the sizing of the Ru capture system.

Second tier data gaps include the determination of the adsorption rates (Gap ID #2, #4, and #6) as these will aid in the determination of the required length of the bed to achieve the desired Ru decontamination factor.

It may be possible to collect all of this initial data in a thermal gravimetric analyzer (TGA)–type system using thin beds containing gram quantities of sorbent or in a deep-bed system in which multiple layers are analyzed. A major challenge in this effort is the generation of an appropriate Ru-containing simulant off-gas stream.

The third tier data needs include the validation of the system DF and length of the MTZ (Gap ID #24 and #25). This will allow further refinement of the design bed length to allow the major portion of the bed to achieve saturation before breakthrough. These tests will require a deep sorbent configuration. The greatest complexity of the test systems described thus far is the increasing complexity of the off-gas supply system required to challenge the sorbent material with more and more prototypic off-gas concentrations. These systems will contain grams to kilograms of sorbent initially. The use of radioactive tracers should also be considered as the number of off-gas components is increased to allow more accurate determination of the co-adsorption capacity of the various Ru adsorbents.

The data gaps for the thermal properties are considered a lower priority at this time. Finally, the data gaps that address the regeneration of the sorbent are currently the lowest priority as it is currently thought that the Ru sorbent is a one-time use material.

Testing of the Ru sorbent and the back-flushable filter should be considered, assuming a suitable Ru and fuel solids simulant feed system can be developed. Testing this integrated subsystem should be completed to demonstrate the Ru DF across the filter and to determine the rate of Ru accumulation on the filter media.

Integrated testing should be conducted that includes the coupling of the Ru abatement system with the iodine capture and tritium capture and recovery systems. This should be completed for both TPT off-gas (TPTOG) stream and dissolver system conditions with a particular focus on the long-term effects of water vapor on the Ru recovery system, as well as the potential accumulation of Ru within the tritium capture systems. Although the direct-dissolution-of- fuel-without-TPT analysis case did not include tritium capture on the dissolver or VOG lines, a recent analysis by Jubin and Spencer (2017) indicates that in the absence of TPT tritium capture may be required on the DOG and VOG systems.

Table 2. Data gaps for ruthenium sorbent evaluations

#	Property	Specific requirements	Approach	Data gap impact	Notes
Metrics for technical performance and physical and chemical characteristics criterion					
1	**Adsorption capacity** *mol/kg sorbent*	Maximum capacity of selected sorbent material as a function of Ru species, temperature, and concentration. - Ru species of interest are may include RuO_4, RuI_4, and $RuBr_4$. - Operating temperature range is 30ºC–600ºC. - Ru concentration ranging from 1×10^{-7} kg/m^3 to 1×10^{-4} kg/m^3. - Gas velocities ranging from 5 to 100 m/min.	Thin-bed testing *Test system must be able to hold a thin bed of sorbent at given temperature and expose sorbent to a generated air stream containing water (and any potentially co-adsorbed species) at varying concentrations within the specified concentration range and gas velocity.*	Equipment size	Only limited data found in the literature on silica gel capacity.
2	**Capture/removal rates for primary species** *mol/kg sorbent/h*	Adsorption rate data for selected sorbent as function of Ru species, temperature, and concentration. Recommended experimental ranges and Ru species are provided in Gap ID #1.	Thin-bed testing *(See Gap ID #1)*	Equipment size	

Table 2. Data gaps for ruthenium sorbent evaluations (continued)

#	Property	Specific requirements	Approach	Data gap impact	Notes
3	**Capacity for other species present in gas stream** *mol/kg sorbent*	Maximum capacity for potential co-absorbed species by selected sorbent material as a function of species, temperature, and concentration. - Species of interest are H_2O, O_2, N_2, HNO_3, CO_2, I, etc. - H_2O with a dew point of −60°C–90°C - HNO_3 concentration of 1,000 ppm - O_2 concentrations from 20% to ~100% - N_2 concentrations of 0%–80% - CO_2 concentrations of 440 ppm - I concentrations up to 0.1 kg/m^3 (10,000 ppm); - Operating temperature range is 30°C–600°C	Thin-bed testing *(See Gap ID #1)*	Equipment size/operational sequencing	
4	**Capture/removal rate for co-absorbed species** *mol/kg sorbent/h*	Adsorption rate data for selected sorbent as function of, co-absorbed species, temperature, and concentration Recommended experimental ranges and potentially co-adsorbing species are provided in Gap ID #3.	Thin-bed testing *(See Gap ID #1)*	Equipment size/operational sequencing	

Table 2. Data gaps for ruthenium sorbent evaluations (continued)

#	Property	Specific requirements	Approach	Data gap impact	Notes
5	**Change in sorbent capacity for Ru in presence of other species present in gas stream** *mol/kg sorbent*	Ru capacity of selected sorbent material for potential adverse effects to Ru capacity in the presence of co-absorbed species function of species, temperature, and concentration. Recommended experimental ranges, Ru species, and potentially co-adsorbing species are provided in Gap IDs #1 and #3.	Thin-bed testing with post-adsorption sorbent analysis *Thin-bed testing (see Gap ID #1) using selected sorbent followed by chemical or gamma analysis of sorbent*	Equipment size	
6	**Change in Ru capture rate in presence of co-absorbed species** *mol/kg sorbent/h*	Ru adsorption rate data for selected sorbent as function of, co-absorbed species, temperature, and concentration. Recommended experimental ranges, Ru species, and potentially co-adsorbing species are provided in Gap IDs #1 and #3.	Thin-bed testing with post-adsorption sorbent analysis *(See Gap ID #5)*	Equipment size	
7	**Selectivity** $(X_a/Y_a)/(X_b/Y_b)$ (unitless) *Where X_a and X_b are mol fractions of species a and b respectively in the adsorbed phase, and Y_a and Y_b are mol fractions of species a and b in the bulk phase*	Derived from data on co-absorption studies shown above.	Thin-bed testing with post-adsorption sorbent analysis *(See Gap ID #5)*	Equipment size/operational sequencing	

Table 2. Data gaps for ruthenium sorbent evaluations (continued)

#	Property	Specific requirements	Approach	Data gap impact	Notes
8	**Sorbent particle density** *kg/m^3*	Density of individual particles in kg/m^3.	Direct measurement or from manufacturer	Equipment size	Data available from Garg Chemical for silica gel.
9	**Sorbent bulk density** *kg/m^3*	Bulk density of bed of sorbent material in typical loading configuration in kg/m^3.	Direct measurement or from manufacturer	Equipment size	
10	**Specific heat capacity** *J/K/kg*	Specific heat capacity of fresh and loaded sorbent over range of operating conditions (30 °C to 600 °C).	Direct measurement of fresh sorbent and loaded sorbent	Heat duty	
11	**Thermal conductivity** *W/m/K*	Thermal conductivity of fresh and loaded sorbent over range of operating conditions (30 °C to 600 °C).	Direct measurement of fresh sorbent and loaded sorbent	Heat duty/operational sequencing	
12	**Radiation stability** *% degradation in capacity, adsorption rate, regeneration time, and MTZ over time as a function of radiation exposure*	Total adsorbed dose for Ru sorbents assume one time use, i.e, no regeneration. Total dose requirement is design dependent. Total dose should factor in both external dose and internal dose.	Irradiation and thin-bed testing *Exposure of the sorbent to both γ and β radiation for a range of total doses. Testing of the sorbent with both thin- and deep-bed testing to characterize radiation effects.* *(Thin-bed testing: See Gap IDs #1 and #5)*	Material life cycle	

Table 2. Data gaps for ruthenium sorbent evaluations (continued)

#	Property	Specific requirements	Approach	Data gap impact	Notes
13	**Mechanical stability** *N/mm (load vs particle diameter)* *μg/kg sorbent loss to gas stream*	Determination of the fines generation and mechanical strength of the sorbent material.	Direct measurement *Measurements of both particle crush strength and abrasion resistance will be required*	Material life cycle	
14	**Thermal stability** *% degradation in capacity over time at selected operating temperature*	Determine the extent of capacity loss due to extended exposure to operating temperature. Determine the extent of capacity loss due to thermal cycling and excursions.	Extended testing with thin-bed analysis. The sorbent should be exposed to the operating temperature for extended periods of time and should experience thermal cycling to simulate regeneration of the sorbent. The capacity should be measured after thermal exposure by thin-bed testing. *(See Gap ID #1)*	Material life cycle	

Table 2. Data gaps for ruthenium sorbent evaluations (continued)

#	Property	Specific requirements	Approach	Data gap impact	Notes
15	**Chemical stability** *% degradation in capacity over time as a function of other species present in gas stream*	Determine the extent of capacity loss from extended exposure to other species present in the off-gas stream.	Extended testing with thin-bed analysis. *Age sorbents for various lengths of time to elevated levels of each of the non-target species expected. Determine Ru capacity and rates of adsorption. Differences should be attributable to aging and should be a function of exposure time and non-target species exposure.*	Material life cycle	Unexpected species present in the gas stream, particularly from upset conditions, impacts the operating life and performance of the sorbent.
16	**Reactivity** *Compatibility as determined by standardized compatibility tables*	Confirmation that any compatibility issues can be avoided through selection of materials of construction, appropriate pretreatment of gas stream, operational envelope, etc.	Direct evaluation.	Material life cycle	This examines the interaction between the sorbent and materials of construction and other gas stream components. This was assumed not to be a problem in the Engineering Evaluation study but was never confirmed.
17	**Regeneration capacity stability** *Number of cycles and Δ mol/kg sorbent*	No. of cycles before degrading to 80% of capacity for the target element.	Extended testing with thin-bed analysis. *Long-term, repeated testing of select sorbent over multiple adsorption/ desorption cycles if sorbent is to be regenerated. N/A if single use.*	Material life cycle	Not currently considered a factor for Ru sorbents as these are most likely one time use.
18	**Desorption rate of Ru** *mol/kg sorbent/h*	Desorption rate as a function of desorption temperature and purge stream gas velocity.	Thin-bed testing. *(See Gap ID #1)*	Equipment size	

Table 2. Data gaps for ruthenium sorbent evaluations (continued)

#	Property	Specific requirements	Approach	Data gap impact	Notes
19	**Desorption of co-adsorbed species** *mol co-adsorbed species retained/m^3 sorbent*	% of co-adsorbed species desorbed during selected regeneration conditions. Recommended experimental ranges and potentially co-adsorbing species are provided in Gap ID #3.	Thin-bed testing. *(See Gap ID #1)*	Material life cycle	
20	**Desorption rate of co-adsorbed species** *mol/kg sorbent/h*	Desorption rate as a function of desorption temperature and purge stream gas velocity. Recommended experimental ranges and potentially co-adsorbing species are provided in Gap ID #3.	Thin-bed testing. *(See Gap ID #1)*	Material life cycle	
21	**Purity of recovered Ru** *mol impurity/kg Ru*	Prescribed by waste treatment operation.	ICP-MS of recovered phase.	Equipment size and waste treatment process design	Not considered because current design is single use.
22	**Cooling time** *h*	Hours to cool from regeneration temperature to adsorbing temperature (30°C)	Derived from heat capacity, thermal conductivity, and bed design.	Heat duty	Readily derived and confirmed during deep-bed testing. Not considered because current design is single use.
			Metrics for system design and performance criterion		
23	**Pressure drop** *Pa/m vs m^2 column*	Size columns and sorbent size for <2.5 kPa pressure drop based on expected gas flow rates.	Direct measurement or scaled derivation.	Operational sequencing/equipment size	Not considered directly in Engineering Evaluation.

Table 2. Data gaps for ruthenium sorbent evaluations (continued)

#	Property	Specific requirements	Approach	Data gap impact	Notes
24	**Decontamination factor (DF)** $[Ru]_{inlet}/[Ru]_{outlet}$ (unitless)	DF data as function of sorbent, Ru species, temperature, concentration, time, and bed depth. Recommended experimental ranges and Ru species are provided in Gap ID #1.	Deep-bed testing with online measurement of inlet and effluent Ru concentrations. *Test system must be able to hold a deep bed of sorbent at given temperature and expose sorbent to a generated air stream containing water (and any potentially co-adsorbed species) at varying concentrations within the specified concentration range and gas velocity. Both inlet and effluent gas streams must be assessed for Ru concentration.*	Equipment size/operational sequencing	This is actually a function of the overall system design but the ability to demonstrate that the assumed DF is achievable is needed for a comparable design.
25	**Length of Mass Transfer Zone (MTZ)** *m*	Length of MTZ as a function of gas velocity, target species concentration, temperature, and presence of co-absorbed species shown to have impact on total capacity or adsorption rate >10%. Recommended experimental ranges and Ru species are provided in Gap ID #1.	Deep-bed tests with bed depth that captures 1.5 times MTZ. Predictions can be made from thin-bed tests and data may also be determined from DF testing using deep beds. *(Deep-bed testing: Gap ID #24).*	Equipment size	

4.2 Tritium recovery systems

Tritium (^3H) present in the off-gas generated by air TPT process will be present as tritiated water (^3H_2O or $^3H^1HO$) and will range in concentration from 1×10^{-7}kg/m^3 to 1×10^{-4} kg/m^3 (0.1–100 ppm). Several commercially available sorbents are suitable for water removal (gas drying) and of, those molecular sieves (MS) 3A, 4A, 5A, etc. are promising candidates for use in a nuclear fuel reprocessing facility because of their robust physical and chemical properties. The Engineering Evaluation found that water removal by 3A MS was relatively well understood in comparison to other treatment operations reviewed, but there were still a number of outstanding data gaps that could substantially impact the unit design. Table 3 summarizes the data gaps and requirements for the tritium capture system. These gaps follow the same structure described in the criteria report but focus primarily on the properties of the sorbent materials and key operational parameters.

The most substantial data gaps limiting the refinement of the tritium removal system design are related to the co-adsorption of other off-gas species, both radioactive species requiring mitigation and other gaseous makeup components, such as CO_2. These gaps include the capacity of 3A MS for other species present in gas stream, the rate of adsorption by 3A MS for other species present in gas stream, the change in sorbent capacity for H_2O in presence of other species present in gas stream, the change in the H_2O capture rate in the presence of co-absorbed species, and the selectivity of 3A MS for H_2O over other potentially co-adsorbing species (Gap ID #3–7). These gaps are most likely to affect equipment size, but they could also affect operational sequencing or material life cycle. Co-adsorption data for the tritium removal system is especially important because of its location in the air TPT treatment train. Ruthenium is likely to be removed before the tritium removal unit. Iodine may be removed to address known co-adsorption issues, but the tritium sorbent will be contacted by a number of other constituents of interest (i.e., CO_2, NO_x, and Kr/Xe). These components could affect not only the size and operation of the tritium removal system, but also the downstream source terms should 3A MS remove substantial quantities of the potentially co-adsorbing species. The regeneration systems and tritium recovery, concentration, and solidification processes could be impacted.

Initially, thin-bed testing to determine the extent to which potentially co-adsorbing species adsorb on 3A MS will be helpful in eliminating some of these species as co-adsorbers, but to date no testing has been conducted on this topic. If any species is identified as co-adsorbing, a combination of both thin- and deep-bed testing will be required to respond to Gaps ID #3–7. With the exception of gathering co-adsorption rate data, the systems required do not represent a substantial challenge in design or fabrication.

The other major data gaps relating to tritium removal by 3A MS are related to desorption of both H_2O and any co-adsorbed species (Gap ID #18–21). The desorption of H_2O has only been characterized under limited conditions. Desorption (or lack thereof) of co-adsorbed species should also be characterized as it may affect material life cycle or equipment sizing. Characterization of the desorption of co-adsorbed species should be done after the identification of co-adsorbed species has been determined. All desorption testing requires the same test equipment that is needed for characterizing H_2O adsorption and co-adsorption of other gaseous species.

The regeneration systems and tritium recovery, concentration, and solidification processes could be impacted by the desorption characteristics of H_2O and any co-adsorbed species. Multicycle testing of the adsorption, desorption, and regeneration coupled with tritium recovery for disposal should be demonstrated in an integrated fashion. This would require a multicolumn system to demonstrate all aspects of the complete process.

Integrated testing that should be conducted includes coupling of the tritium capture and recovery systems and the iodine capture systems with the Ru abatement system. This should be completed for both the TPTOG stream as well as the dissolver system conditions with a particular focus on the long-term effects of water vapor on the Ru recovery system as well as the potential accumulation of Ru within the tritium capture systems. Although the direct dissolution of the fuel without TPT analysis case did not include tritium capture on the dissolver or VOG lines, a recent analysis by Jubin and Spencer (2017) indicates that, in the absence of TPT, tritium capture may be required on the DOG and VOG systems. Of particular importance is the evaluation of the long-term effects of water vapor on the Ru sorbent and the selective recovery of tritium-contaminated water for conversion to a stable waste form.

Table 3. Data needs for tritium sorbents—Air TPT case

#	**Property** *Units*	**Specific requirements**	**Approach**	**Data gap impact**	**Notes**
Metrics for technical performance and physical and chemical characteristics criterion					
1	**Adsorption capacity** *mol/kg*	Maximum capacity of 3A MS as a function of H_2O concentration, temperature, and gas velocity. - H_2O concentration range of interest is 1×10^{-7}kg/m^3 to 1×10^{-4} kg/m^3 (0.1–100 ppm). - Operating temperature ranges of15°C–4°C - Gas velocities ranging from 5 to 100 m/min	Thin-bed testing. *Test system must be able to hold a thin bed of sorbent at given temperature and expose sorbent to a generated air stream containing water (and any potentially co-adsorbed species) at varying concentrations within the specified concentration range and gas velocity.*	Equipment size	Isotherms for 3A MS sorbents are available over a range of operating temperatures and H_2O concentrations. Previous work by the Off-Gas Sigma Team has characterized adsorption of H_2O by 3A MS at feed stream dew points ranging from −10°C to 10°C and sorbent temperature of 30°C (gas velocity 10 m/min).
2	**Capture/removal rates for primary species** *mol/kg sorbent/h*	Adsorption rate data for 3A MS as function of concentration and temperature. Recommended experimental ranges are provided in Gap ID #1.	Thin-bed testing *(See Gap ID #1)*	Equipment size	OGST has characterized adsorption of H_2O by 3A MS at feed stream dew points ranging from −10°C to 10°C and sorbent temperature of 30°C (gas velocity 10 m/min).

Table 3. Data needs for tritium sorbents—Air TPT case (continued)

#	Property *Units*	Specific requirements	Approach	Data gap impact	Notes
3	**Capacity for other species present in gas stream** *mol/kg sorbent*	Maximum capacity for potential co-absorbed species on 3A MS as a function of species and concentration - Species of interest are O_2, CO_2, Xe, Kr, N_2, NO *O_2 (21 vol%)* *CO_2 (~400 ppm)* *Xe (800 ppm)* *Kr (80 ppm)* *N_2 (79 vol%)* *Ar (0.9 vol%)* - Operating temperature ranges of 15°C–40ºC - Gas velocities ranging from 5 to 100 m/min	Thin-bed testing *(See Gap ID #1)*	Equipment size/operational sequencing	
4	**Capture/removal rate for co-absorbed species** *mol/kg sorbent/h*	Adsorption rate data for 3A MS as function of co-absorbed species, their concentration, temperature, and gas velocity. Species of interest and recommended experimental ranges are provided in Gap ID #3.	Thin-bed testing *(See Gap ID #1)*	Equipment size/operational sequencing	

Table 3. Data needs for tritium sorbents—Air TPT case (continued)

#	Property *Units*	Specific requirements	Approach	Data gap impact	Notes
5	**Change in sorbent capacity for H_2O in presence of other species present in gas stream** *mol/kg sorbent*	H_2O capacity of selected sorbent material in the presence of co-absorbed species function of species and concentration, temperature, and gas velocity. Species of interest, their concentrations, and recommended experimental conditions are provided in Gap #3.	Deep-bed testing with condensation of desorbed H_2O. *Test system must be able to hold a deep bed of sorbent at given temperature and expose sorbent to a generated air stream containing water (and any potentially co-adsorbed species) at varying concentrations within the specified concentration range and gas velocity.* *The effluent from the deep bed should be passed through a cold trap to condense H_2O from the effluent stream.* *Analysis of condensate will assess whether H_2O capacity was affected by the presence of other gaseous species.*	Equipment size/operational sequencing	

Table 3. Data needs for tritium sorbents—Air TPT case (continued)

#	Property *Units*	Specific requirements	Approach	Data gap impact	Notes
6	**Change in H_2O capture rate in presence of co-absorbed species** *mol/kg sorbent /h*	H_2O adsorption rate data for 3A MS as function of co-absorbed species, their concentrations, temperature, and gas velocity. Species of interest, their concentrations, and recommended experimental conditions are provided in Gap #3.	Thin-bed testing with time-resolved sorbent analysis *Test system must be able to hold a thin bed of sorbent at given temperature and expose sorbent to a generated air stream containing water (and any potentially co-adsorbed species) at varying concentrations within the specified concentration range and gas velocity.* *The sorbent should be analyzed during the test (or the test should be terminated at discrete time intervals) to resolve the contributions of H_2O (primary species) and co-adsorbed species to the observed weight gain.*	Equipment size/operational sequencing	

Table 3. Data needs for tritium sorbents—Air TPT case (continued)

#	Property *Units*	Specific requirements	Approach	Data gap impact	Notes
7	**Selectivity** $(X_a/Y_a)/(X_b/Y_b)$ (unitless) *Where X_a and X_b are mol fractions of species a and b respectively in the adsorbed phase, and Y_a and Y_b are mol fractions of species a and b in the bulk phase*	Derived from Gap IDs #1–7	Deep-bed testing with condensation of desorbed H_2O. *(See Gap ID #5)*	Equipment size/operational sequencing	
8	**Sorbent particle density** *kg/m^3*	Density of individual particles in kg/m^3	Direct measurement or from manufacturer	Equipment size	Available from manufacturer
9	**Sorbent bulk density** *kg/m^3*	Bulk density of bed of sorbent material in typical loading configuration in kg/m^3.	Direct measurement or from manufacturer	Equipment size	Available from manufacturer
10	**Specific heat capacity** *J/K/kg*	Specific heat capacity of fresh and loaded sorbent as a function of temperature. Temperature range of interest is 15°C–40°C	Direct measurement or from manufacturer	Heat duty	Available from manufacturer
11	**Thermal conductivity** *W/m/K*	Thermal conductivity of fresh and loaded sorbent as a function of temperature. Temperature range of interest is 15°C–40°C	Direct measurement	Heat duty/operational sequencing	Available from manufacturer

Table 3. Data needs for tritium sorbents—Air TPT case (continued)

#	**Property** ***Units***	**Specific requirements**	**Approach**	**Data gap impact**	**Notes**
12	**Radiation stability** *% degradation in capacity, adsorption rate, regeneration time, and MTZ over time as a function of radiation exposure*	Capacity for H_2O, adsorption rate, and regeneration time for 3A MS as a function of adsorbed dose. Total dose requirement is design dependent. Total dose should factor in both external and internal dose.	Irradiation, thin-bed testing, and deep-bed testing *Exposure of the sorbent to both γ and β radiation for a range of total doses. Testing of the sorbent with both thin- and deep-bed testing to characterize radiation effects.* *(Thin-bed testing: See Gap ID #1)* *(Deep-bed testing: See Gap ID #5)*	Material life cycle	
13	**Mechanical stability** *N/mm (load vs particle diameter)* *$\mu g/m^3$ loss to gas stream*	Determination of the fines generation and mechanical strength of the sorbent material.	Direct measurement *Measurements of both particle crush strength and abrasion resistance will be required*	Material life cycle	Degradations in mechanical stability of 3A MS have not been observed in previous OGST testing.

Table 3. Data needs for tritium sorbents—Air TPT case (continued)

#	Property *Units*	Specific requirements	Approach	Data gap impact	Notes
14	**Thermal stability** *% degradation in capacity after exposure to multiple adsorption/regeneration cycles*	Determine the extent of capacity loss due to extended exposure to operating temperature. Determine the extent of capacity loss due to thermal cycling and excursions.	Extended testing with thin-bed analysis. The sorbent should be exposed to the operating temperature for extended periods of time, and should experience thermal cycling to simulate regeneration of the sorbent. The capacity should be measured after thermal exposure by thin-bed testing. *(Thin-bed testing: See Gap ID #1)*	Material life cycle	Degradations in thermal stability of 3A MS have not been observed in previous OGST testing, with up to 3 regeneration cycles.
15	**Chemical stability** *% degradation in capacity over time as a function of other species present in gas stream*	Confirmation that any compatibility issues can be avoided through selection of materials of construction, appropriate pretreatment of gas stream, operational envelope, etc.	Extended testing with thin-bed analysis. *Age sorbents for various lengths of time to elevated levels of each of the non-target species expected. Determine H_2O capacity and rates of adsorption. Differences should be attributable to aging and should be a function of exposure time and non-target species exposure.* *(Thin-bed testing; See Gap ID #1)*	Material life cycle	3A MS sorbent is chemically stable in basic, neutral, and mildly acidic environments.

Table 3. Data needs for tritium sorbents—Air TPT case (continued)

#	**Property** ***Units***	**Specific requirements**	**Approach**	**Data gap impact**	**Notes**
16	**Reactivity** *Compatibility as determined by standardized compatibility tables*	Demonstrated compatibility with all components of gas stream and materials of construction.	Direct evaluation.	Material life cycle	3A MS is compatible with common materials of construction and is chemically stable in basic, neutral, and mildly acidic environments.
17	**Regeneration capacity stability** *% degradation in H_2O capacity per 10 regeneration cycles*	No. of cycles before degrading to 80% of capacity for the target element.	Extended testing with thin-bed analysis. *Long-term, repeated testing of select sorbent over multiple adsorption/ desorption cycles if sorbent is to be regenerated. N/A if single use.* *(Thin-bed testing; See Gap ID #1)*	Material life cycle	OGST has obtained data measuring the H_2O capacity during 3 regeneration cycles and no degradation in capacity was observed.
18	**Desorption rate of H_2O** *mol/kg sorbent/h*	Desorption rate as a function of desorption temperature and purge stream gas velocity.	Thin-bed testing. *(See Gap ID #1)*	Equipment size/operational sequencing	OGST has measured for a desorption temperature of 270°C for a single flow rate.
19	**Desorption of co-adsorbed species** *mol co-adsorbed species retained/ kg sorbent*	% of co-adsorbed species desorbed during selected regeneration conditions. Species of interest, their concentrations, and temperature and gas velocity ranges are found in Gap ID #3.	Thin-bed testing with post-test sorbent analysis. *(See Gap ID #1)*	Equipment size	

Table 3. Data needs for tritium sorbents—Air TPT case (continued)

#	Property *Units*	Specific requirements	Approach	Data gap impact	Notes
20	**Desorption rate of co-adsorbed species** *mol/m^3/h*	Desorption rate of co-adsorbed species as a function of temperature and gas velocity. Species of interest, their concentrations, and temperature and gas velocity ranges are found in Gap ID #3.	Thin-bed testing with time-resolved sorbent analysis. *(See Gap ID #6)*	Material life cycle	
21	**Purity of condensate** *mol impurity/kg ^{3}H-bearing condensate*	The level of impurities tolerated is determined by the waste acceptance criteria for the intended disposition facility.	Deep-bed testing with condensation of desorbed H_2O and condensate analysis.	Equipment size	
22	**Cooling time** *h*	Hours to cool from regeneration temperature (270°C) to adsorbing temperature (30°C).	Derived from heat capacity, thermal conductivity, and bed design.	Heat duty	
		Metrics for system design and performance criterion			
23	**Pressure drop** *Pa/m vs m^2 column*	Pressure drop as a function of column length.	Direct measurement (or scaled derivation).	Equipment size	

Table 3. Data needs for tritium sorbents—Air TPT case (continued)

#	**Property** ***Units***	**Specific requirements**	**Approach**	**Data gap impact**	**Notes**
24	**Decontamination factor (DF)** *$[Isotope]_a/[Isotope]_b$* *$[H_2O]_{inlet}/[H_2O]_{outlet}$* (unitless)	DF data as function of H_2O concentration, time, bed depth and gas velocity. Recommended experimental ranges are found in Gap ID #1. DF requirements will be design dependent.	Deep-bed testing with online measurement of inlet and effluent H_2O concentrations. *Test system must be able to hold a deep bed of sorbent at given temperature and expose sorbent to a generated air stream containing water (and any potentially co-adsorbed species) at varying concentrations within the specified concentration range and gas velocity. Both inlet and effluent gas streams must be assessed for H_2O concentration.*	Equipment size	OGST has collected DF data for a limited condition set.

Table 3. Data needs for tritium sorbents—Air TPT case (continued)

#	**Property** ***Units***	Specific requirements	Approach	Data gap impact	Notes
25	**Length of Mass Transfer Zone (MTZ)** *m*	Length of MTZ as a function of gas velocity, H_2O concentration, and presence of co-absorbed species shown to have impact on total capacity or adsorption rate > 10%. Recommended experimental ranges are found in Gap ID #1.	Deep-bed testing with online measurement of inlet and effluent H_2O concentrations. *Test system must be able to hold a deep bed of sorbent at given temperature and expose sorbent to a generated air stream containing water (and any potentially co-adsorbed species) at varying concentrations within the specified concentration range and gas velocity.*	Equipment size	OGST has collected MTZ data for a limited condition set.

4.3 Iodine recovery systems

Radioactive iodine capture could be one of the more challenging volatile or semi-volatile radionuclides requiring abatement. ^{129}I could be present in multiple off-gas streams (including the DOG, VOG, and air TPT streams), may present as different gas species, and may be present at concentrations that could vary by orders of magnitude. Relatively high iodine removal efficiencies are expected to be required. The different expected off-gas streams could have flow rates that vary by orders of magnitude from each other. These conditions dictate a highly efficient iodine capture technology that can be adapted in different sorption designs tailored for different off-gas systems with different gas flow rates and compositions. Current Department of Energy R&D activities focus on solid silver-based iodine sorbents. Iodine adsorption research is currently being performed using both thin-bed and deep-bed test systems at Oak Ridge National Laboratory (ORNL) and Idaho National Laboratory, and the data from that testing and past work was reflected in the Engineering Evaluation. This experience base and data are accounted for by the authors in the preparation of the iodine adsorption data gap tables. Although a substantial amount of data is available relating to iodine abatement by silver sorbents, the majority of this data has been collected at conditions relevant to the DOG, and iodine removal from the VOG or air TPT streams is notably less understood.

Table 4 summarizes the data gaps for iodine adsorption processes for DOG systems. Table 5 summarizes data gaps for the VOG and air TPTOG systems, which are expected to have iodine concentrations about two orders of magnitude lower than in DOG systems. The extremely dilute iodine concentrations of the VOG and air TPT present a substantial challenge for iodine abatement compared to the DOG. The VOG and air TPT streams may have similar iodine concentrations, but the iodine speciation is very different, with iodine in the air TPTOG expected to be primarily elemental I_2 and in the VOG expected to be a mixture of various organic iodides. The need to achieve high removal efficiencies for such low-concentration gas streams, together with the much higher expected VOG system flow rate and iodine species, could impose different sorbent system design and operating requirements. These different performance requirements dictate some differences in how R&D activities relating to iodine capture from VOG and air TPT streams need to be tailored compared to R&D on iodine capture from DOG streams.

Recommended experimental systems for both DOG and VOG/air TPT are primarily thin- and deep-bed test systems. Thin-bed testing is beneficial for determining the iodine capture capacity and rate because these parameters can be measured in real time when the thin-bed system is inside a TGA. Deep-bed testing is necessary to determine capture efficiency and the length of the MTZ. However, the lower expected iodine concentrations in VOG and air TPTOG streams dictates much longer-duration testing, which challenges the ability to operate both thin-bed TGA and deep-bed test systems stably to produce the needed data. Further, extremely dilute iodine concentrations may limit the applicability of thin-bed and TGA testing for VOG/air TPT experiments.

The highest priority data gaps for iodine recovery systems are the DF (Gap #24) and length of the MTZ (Gap #25). Values for DF as well as estimates for the length of the MTZ are available for some DOG conditions, but there is little relevant information available for VOG and air TPT systems. The MTZ depth and the DF were explicitly discussed in the Engineering Evaluation as being of high importance in the off-gas abatement system design process. Determination of these parameters in the cases of VOG and air TPT could require tests on the time order of months or years.

A similar priority data need is the continuing collection of basic adsorption performance data for VOG and air TPT systems (Gap IDs #1–2). This information has been collected for DOG systems over selected experimental ranges. To accurately resolve Gaps #1–2 for VOG/air TPT, longer term tests will need to be carried out, and a prototype of this testing was initiated in fiscal year 2017 at ORNL.

The next tier of data gaps is the determination of the impact of co-adsorbed species on iodine capture (Gaps #3–7). This includes the concentration of contaminants expected to adsorb onto the iodine capture material and determination of whether those species would depress the iodine capture rate or capacity. This information would require thin- and deep-bed tests. Gaps #3–7 have been investigated for DOG systems, but almost no information is available for VOG/air TPT systems.

The third tier of data gaps is the desorption behavior both of iodine and co-adsorbed species (Gaps #19–21). There will be no regeneration of sorbent material, but physisorbed iodine will need to be removed before sorbent changeout. This data gap applies to all iodine abatement systems.

The data gaps regarding the thermal properties are considered a lower priority at this time. Finally, the data gaps that address the regeneration of the sorbent are currently the lowest priority because the iodine sorbent is currently considered a one-time use material.

Table 4. Data gaps for DOG iodine sorbent evaluations

#	**Property** ***Units***	**Specific requirements**	**Approach**	**Data gap impact**	**Notes**
Metrics for technical performance and physical and chemical characteristics criterion					
1	**Adsorption Capacity** *mol/kg sorbent*	Maximum capacity of selected sorbent as a function of iodine species and their concentrations, temperature, and gas velocity. - Iodine species of interest for DOG may include I_2, CH_3I, HI, and others. - Operating temperatures ranging from 125ºC to 175ºC. - Iodine concentrations ranging from 1×10^{-5} kg/m3 to 1×10^{-3} kg/m^3 (1 to 100 ppm). - Gas velocities ranging from 5 to 100 m/min.	Thin-bed tests using selected sorbent; and deep-bed tests for conditions at time of breakthrough. *Test systems must be able to hold thin and deep beds of sorbent at given temperature and expose sorbent to a generated air stream containing water (and any potentially co-adsorbed species) at varying concentrations within the specified concentration range and gas velocity.* *Sorbents should be analyzed after testing to confirm iodine loading*	Equipment size	Capacity data exist for AgZ, and Ag-Aerogel at various temperatures, typically at 10 m/min. gas velocity.
2	**Capture/removal rates for primary species** *mol/kg sorbent/h*	Adsorption rate data for selected sorbent as function iodine species and their concentrations, temperature, and gas velocity. Recommended experimental ranges and iodine species are found in Gap ID #1.	Thin-bed testing *(See Gap ID #1)*	Equipment size	Data exist for AgZ, Ag-Aerogel, and AgA at various temperatures, typically at 10 m/min. gas velocity.

Table 4. Data gaps for DOG iodine sorbent evaluations (continued)

#	Property *Units*	Specific Requirements	Approach	Data Gap Impact	Notes
3	**Capacity for other species present in gas stream** *mol/kg sorbent*	Maximum capacity for potential co-absorbed species by selected sorbent material as a function of species, temperature, and concentration. - Species of interest for DOG may include H_2O, HNO_3, CO_2, Cl, and Br, *H_2O with a dew point of 0 to 40°C;* *HNO_3 concentrations from 0 to 1000 ppm;* *CO_2 concentrations from 0 to 440 ppm;* *Cl concentrations dependent on its plant acid concentration* *Br concentrations from 1-100 ppm* - Operating temperatures ranging from 125ºC to 175ºC. - Gas velocities ranging from 5 to 100 m/min.	Thin-bed testing *(See Gap ID #1)*	Equipment size	Information is available for water and NO_x from thin-bed tests. Additional limited data are available from the Coupled End-to-End Research Project (CETE).
4	**Capture/removal rate for co-absorbed species** *mol/kg sorbent/h*	Adsorption rate data for selected sorbent as function of co-absorbed species, their concentrations, temperature, and gas velocity. Recommended experimental ranges and potentially co-adsorbing species are provided in Gap ID #3	Thin-bed testing *(See Gap ID #1)*	Equipment size	Information is available for water and NO_x from thin-bed tests. Additional limited data are available from the CETE.

Table 4. Data gaps for DOG iodine sorbent evaluations (continued)

#	Property *Units*	Specific Requirements	Approach	Data Gap Impact	Notes
5	**Change in sorbent capacity for iodine in presence of other species present in gas stream** *mol/kg sorbent*	Iodine capacity of selected sorbent material in the presence of co-absorbed species as a function of species, temperature, and concentration. Recommended experimental ranges, iodine species, and potentially co-adsorbing species are provided in Gap IDs #1 and 3.	Thin-bed testing with post-adsorption sorbent analysis. *Thin-bed testing (see Gap ID #1) using selected sorbent followed by chemical or gamma analysis of sorbent*	Equipment size	Information is available for water and NO_x from thin-bed tests. Additional limited data are available from the CETE.
6	**Change in iodine capture rate in presence of co-absorbed species** *mol/kg sorbent /h*	Iodine adsorption rate data for selected sorbent as function of co-absorbed species, temperature, and concentration. Recommended experimental ranges, iodine species, and potentially co-adsorbing species are provided in Gap IDs #1 and 3.	Thin-bed testing. *(See Gap ID #1)*	Equipment size	Information is available for water and NO_x from thin-bed tests. Additional limited data are available from the CETE.
7	**Selectivity** $(X_a/Y_a)/(X_b/Y_b)$ (unitless) *Where X_a and X_b are mol fractions of species a and b respectively in the adsorbed phase, and Y_a and Y_b are mol fractions of species a and b in the bulk*	Derived from Gap ID #3–6.	Thin-bed testing with post-adsorption sorbent analysis. *(See Gap ID #5)*	Equipment size	Derived from data on co-absorption studies shown above. Limited data are available from the CETE. Should include information on general co-adsorbents (such as NO_x and water), or with tramp halogens (chlorine, bromine, fluorine).

Table 4. Data gaps for DOG iodine sorbent evaluations (continued)

#	**Property** ***Units***	**Specific Requirements**	**Approach**	**Data Gap Impact**	**Notes**
	phase				
8	**Sorbent particle density** *kg/m^3*	Density of individual particles in kg/m^3.	Direct measurement or from manufacturer.	Equipment size	
9	**Sorbent bulk density** *kg/m^3*	Bulk density of bed of sorbent material in typical loading configuration in kg/m^3.	Direct measurement or from manufacturer.	Equipment size	
10	**Specific heat capacity** *J/K/kg*	Specific heat capacity of fresh and loaded sorbent from 25°C–300°C	Direct measurement of fresh sorbent and loaded sorbent.	Heat duty	
11	**Thermal conductivity** *W/m/K*	Thermal conductivity of fresh and loaded sorbent from 25°C–300°C.	Direct measurement of fresh sorbent and loaded sorbent.	Heat duty	
12	**Radiation stability** *% degradation in capacity, adsorption rate, regeneration time, and MTZ over time as a function of radiation exposure*	Capacity for iodine and iodine adsorption rate as a function of adsorbed dose. Total dose requirement is design dependent. Total dose should factor in both external dose and internal dose.	Irradiation and thin-bed testing. *Exposure of the sorbent to both γ and β radiation for a range of total doses. Testing of the sorbent with both thin- and deep-bed testing to characterize radiation effects.* *(Thin-bed testing: See Gap IDs #1 and #5)*	Material life cycle	
13	**Mechanical stability** *N/mm (load vs particle diameter)*	Determination of the fines generation and mechanical strength of the sorbent material.	Direct measurement. *Measurements of both particle crush strength and abrasion resistance*	Material life cycle	Previous tests have indicated that Ag-Aerogel could generate fines under some off-gas conditions; AgZ appears to be stable.

Table 4. Data gaps for DOG iodine sorbent evaluations (continued)

#	Property *Units*	Specific Requirements	Approach	Data Gap Impact	Notes
	μg/m³ loss to gas stream		*will be required*		
14	**Thermal stability** *% degradation in capacity over time at selected operating temperature*	Determine the extent of capacity loss due to extended exposure to operating temperature. Determine the extent of capacity loss due to thermal excursions.	Thin-bed testing. *(See Gap ID #1)*	Material life cycle	Previous tests have not resulted in appreciable thermal instability over time intervals for AgZ lifetime in DOG systems
15	**Chemical stability** % degradation in capacity over time as a function of other species present in gas stream	Determine the degradation in sorption performance (capacity, rate, etc.) as a function of contaminants in the gas feed system (e.g., NO_x, tramp halogens, etc.).	Thin-bed testing. *(See Gap ID #1)*	Material life cycle	
16	**Reactivity** *Compatibility as determined by standardized compatibility tables*	Confirmation that any compatibility issues can be avoided through selection of materials of construction, appropriate pretreatment of gas stream, operational envelope, etc.	Direct evaluation.	Material life cycle	AgZ appears to be compatible with all elements of the system.
17	**Regeneration capacity stability** *% initial capacity vs. cycle*	% degradation as a function of regeneration temperature and duration. This is not applicable for Ag-based iodine adsorbents; one-time use only.	N/A	N/A	Not currently considered a factor for iodine sorbents as these are most likely one time use.
18	**Desorption rate of iodine during post loading purge** *mol/kg sorbent/h*	Desorption of iodine as a function of operating temperature and purge gas velocity. - Operating temperatures ranging from 125ºC to	Thin-bed testing. *(See Gap ID #1)*	Operational sequencing	Some data are available for existing AgZ and Ag-Aerogel sorbent from thin- and deep-bed testing

Table 4. Data gaps for DOG iodine sorbent evaluations (continued)

#	Property *Units*	Specific Requirements	Approach	Data Gap Impact	Notes
		175ºC. - Gas velocities ranging from 5 to 100 m/min.			
19	**Desorption of co-adsorbed species** *mol co-adsorbed species retained/ kg sorbent*	% of co-adsorbed species desorbed during selected regeneration conditions. Recommended experimental ranges and potentially co-adsorbing species are provided in Gap ID #3.	Thin- and deep-bed testing. *(See Gap ID #1)*	Equipment size/material life cycle	
20	**Desorption rate of co-adsorbed species** *mol/kg sorbent /h*	Desorption rate as a function of desorption temperature and purge stream gas velocity. Recommended experimental ranges and potentially co-adsorbing species are provided in Gap ID #3.	Thin- and deep-bed testing. *(See Gap ID #1)*	Material life cycle	
21	**Purity of desorbed iodine streams** *ppm or % iodine*	Prescribed by waste treatment operation.	Purge of deep beds used in previous tests with gas analysis.	Equipment size and waste treatment process design	Sizing of downstream processes
22	**Cooling time** *h*	Hours to cool from pretreatment temperature (270°C) to adsorbing temperature (150°C) and to ambient temperature after purge.	Derived from heat capacity, thermal conductivity, and bed design.	N/A	Readily derived and confirmed during deep-bed testing.
Metrics for system design and performance criterion					
23	**Pressure drop** *Pa/m vs m^2 column*	Size columns and sorbent size for <2.5 kPa pressure drop based on expected gas flow rates	Direct measurement or scaled derivation	Operational sequencing/equipment size	
24	**Decontamination factor (DF)**	DF data as function of sorbent, iodine species, their concentrations, temperature, and	Deep-bed testing with online measurement of inlet and effluent iodine	Equipment size	DFs exist for I_2 and methyl iodide adsorption on AgZ and Ag-Aerogel for selected, limited test conditions; have been for reported for higher-

Table 4. Data gaps for DOG iodine sorbent evaluations (continued)

#	**Property** ***Units***	**Specific Requirements**	**Approach**	**Data Gap Impact**	**Notes**
	$[I]_{inlet}/[I]_{outlet}$ (unitless)	bed depth. Recommended experimental ranges and iodine species are provided in Gap ID #1.	concentrations. *Test system must be able to hold a deep bed of sorbent at given temperature and expose sorbent to a generated air stream containing water (and any potentially co-adsorbed species) at varying concentrations within the specified concentration range and gas velocity. Both inlet and effluent gas streams must be assessed for iodine concentration.*		concentration (~ 1×10^3 ppm) iodine systems
25	**Length of Mass Transfer Zone (MTZ)** *m*	Length of MTZ as a function of gas velocity, target species concentration, temperature, and presence of co-absorbed species shown to have impact on total capacity or adsorption rate >10%. Recommended experimental ranges and iodine species are provided in Gap ID #1.	Deep-bed tests with bed depth that captures 1.5 times MTZ. Predictions can be made from thin-bed tests and data may also be determined from DF testing using deep beds. *(Deep-bed testing: Gap ID #24).*	Equipment size	Estimates of length of MTZ exist for I_2 and methyl iodide adsorption on AgZ and Ag-Aerogel for selected, limited test conditions.

Table 5. Data gaps for iodine sorbent evaluations for air TPT and VOG gas streams

#	Property	Specific requirements	Approach	Data gap impact	Notes
Metrics for technical performance and physical and chemical characteristics criterion					
1	**Adsorption capacity** *mol/kg sorbent*	Maximum capacity of selected sorbent as a function of iodine species and their concentrations, temperature, and gas velocity. - Iodine species of interest for VOG may include I_2, CH_3I, and various organic iodides. For air TPT iodine is expected to be I_2. - Operating temperatures ranging from 25ºC to 175ºC. - Iodine concentrations ranging from 1×10^{-55} kg/m^3 to 1×10^{-7} kg/m^3 (from 0.1 to 1 ppm). - Gas velocities ranging from 5 to 100 m/min.	Extended-duration thin-bed or hybrid thin-deep bed testing. *Test system must be able to hold a thin or deep bed of sorbent at given temperature and expose sorbent to a generated air stream containing water (and any potentially co-adsorbed species) at varying concentrations within the specified concentration range and gas velocity.* *Sorbents should be analyzed after testing to confirm iodine loading*	Equipment size	ORNL has an extended test in progress that is designed to determine the saturation concentration of I_2 on AgZ under VOG conditions
2	**Capture/removal rates for primary species** *mol/kg sorbent /h*	Adsorption rate data for selected sorbent as function of iodine species, temperature, and concentration. Recommended experimental ranges and iodine species are provided in Gap ID #1.	Extended-duration thin-bed or hybrid thin-deep bed testing (See Gap ID #1) with the capability to perform online or intermittent sorbent analysis.	Equipment size	

Table 5. Data gaps for iodine sorbent evaluations for air TPT and VOG gas streams (continued)

#	Property	Specific requirements	Approach	Data gap impact	Notes
3	**Capacity for other species present in gas stream** *mol/kg sorbent*	Maximum capacity for potential co-absorbed species by selected sorbent material as a function of species, temperature, and concentration. - Species of interest for VOG may include H_2O, HNO_3, CO_2, Cl, and Br, *H_2O with a dew point of 0 to 40°C;* *HNO_3 concentrations from 0 to 1,000 ppm;* *CO_2 concentrations from 0 to 440 ppm;* *Cl concentrations dependent on purity of plant acid* *Br concentrations from 0 to 1 ppm* - Operating temperatures ranging from 125ºC to 175ºC. - Gas velocities ranging from 5 to 100 m/min.	Extended-duration thin-bed or hybrid thin-deep bed testing. *(See Gap ID #1)*	Equipment size	Information is available for water and NO_x from thin-bed tests for a limited condition set. Additional limited data are available from the CETE experiment.
4	**Capture/removal rate for co-absorbed species** *mol/kg sorbent/h*	Adsorption rate data for selected sorbent as function of co-absorbed species, their concentrations, temperature, and gas velocity. Recommended experimental ranges and potentially co-adsorbing species are provided in Gap ID #3.	Extended-duration thin-bed or hybrid thin-deep bed testing (See Gap ID #1) with the capability to perform online or intermittent sorbent analysis.	Equipment size	
5	**Change in sorbent**	Iodine capacity of selected	Extended-duration thin-	Equipment size	

Table 5. Data gaps for iodine sorbent evaluations for air TPT and VOG gas streams (continued)

#	Property	Specific requirements	Approach	Data gap impact	Notes
	capacity for iodine in presence of other species present in gas stream *mol/kg sorbent*	sorbent material in the presence of co-absorbed species as a function of species, temperature, and concentration. Recommended experimental ranges, iodine species, and potentially co-adsorbing species are provided in Gap IDs #1 and 3.	bed or hybrid thin-deep bed testing (See Gap ID #1) with post-adsorption sorbent analysis.		
6	**Change in iodine capture rate in presence of co-absorbed species** *mol/kg sorbent /h*	Iodine adsorption rate data for selected sorbent as function of co-absorbed species, temperature, and concentration. Recommended experimental ranges, iodine species, and potentially co-adsorbing species are provided in Gap IDs #1 and 3.	Extended-duration thin-bed or hybrid thin-deep bed testing (See Gap ID #1) with the capability to perform online or intermittent sorbent analysis.	Equipment size	
7	**Selectivity** $(X_a/Y_a)/(X_b/Y_b)$ (unitless) *Where* X_a *and* X_b *are mol fractions of species a and b respectively in the adsorbed phase, and* Y_a *and* Y_b *are mol fractions of species a and b in the bulk phase*	Derived from Gap ID #3–6.	Thin-bed testing with post-adsorption sorbent analysis. *(See Gap ID #5)*	Equipment size	Derived from data on co-absorption studies shown above. Should include information on general co-adsorbents (such as NO_x and water), or with tramp halogens (chlorine, bromine, fluorine).
8	**Sorbent particle density** *kg/m³*	Density of individual particles in kg/m³.	Direct measurement or from manufacturer.	Equipment size	

Table 5. Data gaps for iodine sorbent evaluations for air TPT and VOG gas streams (continued)

#	Property	Specific requirements	Approach	Data gap impact	Notes
9	**Sorbent bulk density** *kg/m^3*	Bulk density of bed of sorbent material in typical loading configuration in kg/m^3.	Direct measurement or from manufacturer.	Equipment size	
10	**Specific heat capacity** *J/K/kg*	Specific heat capacity of fresh and loaded sorbent from 25°C–300°C.	Direct measurement of fresh sorbent and loaded sorbent.	Heat duty	
11	**Thermal conductivity** *W/m/K*	Thermal conductivity of fresh and loaded sorbent from 25°C–300°C.	Direct measurement of fresh sorbent and loaded sorbent.	Heat duty	
12	**Radiation stability** *% degradation in capacity, adsorption rate, regeneration time, and MTZ over time as a function of radiation exposure*	Capacity for iodine and iodine adsorption rate as a function of adsorbed dose. Total dose requirement is design dependent. Total dose should factor in both external dose and internal dose.	Irradiation and thin-bed testing. *Exposure of the sorbent to both γ and β radiation for a range of total doses. Testing of the sorbent with both thin- and deep-bed testing to characterize radiation effects.* *(Thin-bed testing: See Gap IDs #1 and #5)*	Material life cycle	Information likely necessary as beds are online for very long periods of time (1 year) and may be subject to external dose (design dependent).
13	**Mechanical stability** *N/mm (load vs particle diameter)* *µg/m^3 loss to gas stream*	Determination of the fines generation and mechanical strength of the sorbent material.	Direct measurement. *Measurements of both particle crush strength and abrasion resistance will be required*	Material life cycle	Previous tests have indicated that Ag-Aerogel could generate fines under VOG conditions; AgZ appears to be stable over that timeframe.
14	**Thermal stability** *% degradation in*	Determine the extent of capacity loss due to extended exposure to operating temperature. Determine	Thin-bed testing. *(See Gap ID #1)*	Material life cycle	Previous tests have not resulted in appreciable observed thermal instability over time intervals for AgZ lifetime in DOG systems. VOG/TPTOG

Table 5. Data gaps for iodine sorbent evaluations for air TPT and VOG gas streams (continued)

#	Property	Specific requirements	Approach	Data gap impact	Notes
	capacity over time at selected operating temperature	the extent of capacity loss due to thermal excursions.			will be online much longer.
15	**Chemical stability** % degradation in capacity over time as a function of other species present in gas stream	Determine the degradation in sorption performance (capacity, rate, etc.) as a function of contaminants in the gas feed system (e.g., NO_x, tramp halogens, etc.).	Thin-bed testing. *(See Gap ID #1)*	Material life cycle	Aging tests have shown impact for both AgZ and Ag-Aerogel.
16	**Reactivity** *Compatibility as determined by standardized compatibility tables*	Confirmation that any compatibility issues can be avoided through selection of materials of construction, appropriate pretreatment of gas stream, operational envelope, etc.	Direct evaluation.	Material life cycle	AgZ appears to be compatible with all elements of the system.
17	**Regeneration capacity stability** *% initial capacity vs. cycle*	% degradation as a function of regeneration temperature and duration. This is not applicable for Ag-based iodine adsorbents; one-time use only.	N/A	N/A	
18	**Desorption rate of iodine during post loading purge** *mol/kg sorbent /h*	Desorption of iodine as a function of operating temperature and purge gas velocity. - Operating temperatures ranging from 125ºC to 175ºC. - Gas velocities ranging from 5 to 100 m/min.	Thin-bed testing. *(See Gap ID #1)*	Operational sequencing	Some data are available for existing AgZ and Ag-Aerogel sorbent from thin-bed and deep-bed testing.
19	**Desorption of co-adsorbed species** *mol co-adsorbed species retained/kg sorbent*	% of co-adsorbed species desorbed during selected regeneration conditions. Recommended experimental ranges and potentially co-	Thin- and deep-bed testing. *(See Gap ID #1)*	Equipment size/material life cycle	

Table 5. Data gaps for iodine sorbent evaluations for air TPT and VOG gas streams (continued)

#	Property	Specific requirements	Approach	Data gap impact	Notes
		adsorbing species are provided in Gap ID #3.			
20	**Desorption rate of co-adsorbed species** *mol/kg sorbent /h*	Desorption rate as a function of desorption temperature and purge stream gas velocity. Recommended experimental ranges and potentially co-adsorbing species are provided in Gap ID #3.	Thin- and deep-bed testing. *(See Gap ID #1)*	Material life cycle	
21	**Purity of desorbed iodine streams** *ppm or % iodine*	Prescribed by waste treatment operation.	Purge of deep beds used in previous tests with gas analysis.	Equipment size and waste treatment process design	Sizing of downstream processes.
22	**Cooling time** *h*	Hours to cool from pretreatment temperature (270°C) to adsorbing temperature (150°C) and to ambient temperature after purge.	Derived from heat capacity, thermal conductivity, and bed design.	N/A	
Metrics for system design and performance criterion					
23	**Pressure drop** *Pa/m vs m² column*	Size columns and sorbent size for <2.5 kPa pressure drop based on expected gas flow rates.	Direct measurement or scaled derivation.	Operational sequencing/equipment size	
24	**Decontamination factor (DF)** $[I]_{inlet}/[I]_{outlet}$ (unitless)	DF data as function of sorbent, iodine species, their concentrations, temperature, and bed depth. Recommended experimental ranges and iodine species are provided in Gap ID #1.	Deep-bed testing with online measurement of inlet and effluent iodine concentrations. *Test system must be able to hold a deep bed of sorbent at given temperature and expose sorbent to a generated air stream containing water (and any potentially co-adsorbed species) at*	Equipment size	DFs exist for I_2 and methyl iodide adsorption on AgZ and Ag-Aerogel for selected, limited test conditions.

Table 5. Data gaps for iodine sorbent evaluations for air TPT and VOG gas streams (continued)

#	Property	Specific requirements	Approach	Data gap impact	Notes
			varying concentrations within the specified concentration range and gas velocity. Both inlet and effluent gas streams must be assessed for iodine concentration.		
25	**Length of Mass Transfer Zone (MTZ)** *m*	Length of MTZ as a function of gas velocity, target species concentration, temperature, and presence of co-absorbed species shown to have impact on total capacity or adsorption rate >10%. Recommended experimental ranges and iodine species are provided in Gap ID #1.	Deep-bed tests with bed depth that captures 1.5 times MTZ. Predictions can be made from thin-bed tests and data may also be determined from DF testing using deep beds. *(Deep-bed testing: Gap ID #24).*	Equipment size	Estimates of length of MTZ exist for I_2 and methyl iodide adsorption on AgZ and Ag-Aerogel for selected, limited test conditions.

4.4 NO_x recovery systems

As shown in Figures 1 and 3, the NO_x recovery system is downstream of the iodine trapping system. At this point the off-gas streams have been decontaminated of Ru, ^3H, and iodine to meet requirements, so only trace amounts of these species remain. The primary species arising from fuel dissolution and TPT (when TPT is used) and entering the NO_x system are NO_x (consisting of N_2O, NO, and NO_2), CO, CO_2, Xe, Kr, and the carrier gas (e.g., components of air including N_2, O_2, CO_2, Ar, H_2O, etc.). Vapors of nitric and nitrous acids will also be present in low concentrations. Traces of organic species arising from dissolution may be present. These organics arise from the recycle acid stream where small amounts of organics from solvent extraction operations dissolve or are entrained in the aqueous phase. Some of these organics may have become iodated during dissolution, and stripping of the iodine from the organic molecule in the AgZ treatment step could produce alcohols, ethers, or other organic species.

NO_x recovery systems often scrub the gas phase with water or nitric acid to recover the nitrogen oxides as nitric acid. The acid is recycled and reused within the plant. In the late 1970s and early 1980s, there was a significant effort to better understand the chemical processes (Counce and Groenier 1978) and to develop methods to design sieve-plate (Counce 1978; Counce and Perona 1979a, 1979b and 1980), bubble cap (Counce 1979), and packed (Counce 1980) scrubbing towers in nuclear fuel reprocessing plants. The presence of oxygen is necessary to improve the scrubbing efficiency of NO and N_2O, converting them in situ to NO_2. Fractional recovery of NO_x is variable with tower height (number of theoretical equilibrium stages), is limited to equilibrium vapor pressures above the fed scrubbing solution, and is usually set by optimization of the capital and operating costs. Removal of residual trace amounts of NO_x can be implemented using zeolite-based MS, but this would seem unnecessary because trace amounts are expected to be captured in the CO_2 recovery system (Section 4.5).

The CO, CO_2, inert components of the carrier gas, and the noble gases are sparingly soluble in the acidic aqueous scrubbing solution and will pass to the next step in the off-gas treatment train. Organic compounds will be the more troublesome components entering the NO_x recovery system. How these organics behave is mostly unknown, and there is a need for study to ascertain if such compounds will cause safety or operational problems. For example, a portion of these organics may be converted to organic nitrates or acids that could collect in the aqueous systems. Accumulation of oil-like nitrates is a potential safety issue. Many organic acids are volatile and may be transported to the CO_2 recovery system. Residual trace iodine entering the system may be converted to an iodate, precipitate, and accumulate in the equipment.

Although NO_x scrubbers are considered relatively standard industrial equipment, there appears to be limited experience in the design and sizing of this equipment within the reprocessing community, and there is a declining experience base of staff with experience in scrubber operation. A small pilot facility to support the collection of additional design data and to train staff is needed. Key data that should be of the highest priority is the absorption capacities for both the species of interest and the residual iodine and tritium that may breakthrough the primary abatement systems (Table 6, Gap ID #1–7). It is envisioned that the recovered acid would be recycled within the plant, so the long-term accumulation and potential recycle rates are of interest. Iodine captured in the NO_x scrubber and recycled back to the dissolver would have an additional opportunity to be captured in the DOG iodine sorbent beds, but if recycled back to the acid recovery system, the iodine could be spread to elsewhere within the facility. Little $^{14}CO_2$ co-absorption is expected but should be confirmed.

Table 6. Data gaps for NO_x absorber evaluations

#	Property *Units*	Specific requirements	Approach	Data gap impact	Notes
Metrics for technical performance and physical and chemical characteristics criterion					
1	**Absorption Capacity** *mol/m³ absorbent liquid*	Maximum capacity of scrub solution as a function of NO_x species, their concentrations, temperature, and gas velocity. - NO_x species of interest include NO_2, NO, and N_2O - NO concentration of 4×10^{-4} to 4×10^{-3} kg/m³ (300 to 3000 ppm) - NO_2 1.7×10^{-3} to 1.7×10^{-2} kg/m³ (900–9,000 ppm) - N_2O concentration not known - Gas velocities from 0.18 to 10 m/min - Operating temperature range is −10°C–50°C	Chemical equilibrium modeling and detailed scrubber design; demonstrated in pilot-scale tests.	Equipment size, discharge nitric acid stream flow rate and composition.	Design methodologies have been reported and should be re-evaluated.
2	**Capture removal rates for primary species** *mol/m³ absorbent liquid /h*	Adsorption rate of scrub solution as a function of NO_x species, their concentrations, temperature, and gas velocity. Recommended experimental ranges and NO_x species are provided in Gap ID #1.	Chemical modeling and in situ measurement of HNO_2 concentrations, and changes in concentrations as function of reagents (e.g., O_2 sparge, H_2O_2) and operating conditions.	Equipment size, design, and operating conditions. Design and operation of the downstream CO_2 scrubber.	NO and N_2O scrubbing can be slower; causing current designs to be only 55% to 95% efficient for total NO_x scrubbing. Need to determine effect of HNO_2 concentration and methods to reduce its effect or convert it to HNO_3.

Table 6. Data gaps for NO_x absorber evaluations (continued)

#	Property *Units*	Specific requirements	Approach	Data gap impact	Notes
3	**Capacity for other species present in gas stream** *mol/m³ absorbent liquid*	Maximum capacity for potential co-absorbed species as a function of species, their concentrations, temperature, and gas velocity. Co-adsorbing species may include CO_2, CO, I_2, and organic acids. - Gas velocities of 0.18 to 10 m/min - Operating temperature range is -10°C to 50°C - CO_2 7.7×10^{-4} kg/m³ (400 ppm) - CO not known but not expected to be important. - I_2 8.7×10^{-8} kg/m³ (0.01 ppm) - Organic specie concentrations not known	Column scrubber tests and/or column engineering models. Tests with steam stripping, H_2O_2 addition or other techniques. Identification of organics entering system and conversion to organic nitrates and/or acids.	Equipment size/operational sequencing. Safety issues and degradation of scrubbing efficiency.	Of particular interest: Iodine retention and/or accumulation; escape of less easily scrubbed NO; and disposition of organic compounds, solubility limits.
4	**Capture/removal rate for co-absorbed species** *Mol/m³ absorbent liquid /h*	Adsorption rate data for selected sorbent as function of co-absorbed species, temperature, and concentration Recommended experimental ranges and potentially co-adsorbing species are provided in Gap ID #3.	Calculation from chemical data bases, laboratory studies.	Equipment size/operational sequencing.	Concentrations of other gas species in the DOG may be low enough to limit impact of co-absorbed species on NO_x scrubbing.

Table 6. Data gaps for NO_x absorber evaluations (continued)

#	Property *Units*	Specific requirements	Approach	Data gap impact	Notes
5	**Change in sorbent capacity for NO_x in presence of other species present in gas stream** *mol/m³ absorbent liquid*	NO_x capacity accounting for potential adverse effects from co-absorbed species as a function of species, temperature, and concentration. Recommended experimental ranges, NO_x species, and potentially coasorbing species are provided in Gap IDs #1 and #3.	Calculation from chemical data bases, laboratory studies.	Equipment size	Concentrations of other gas species in the DOG may be low enough to limit impact of co-absorbed species on NO_x scrubbing.
6	**Change in NO_x capture rate in presence of co-absorbed species** *mol/m³ absorbent liquid /h*	NO_x adsorption rate data for selected sorbent as function of co-absorbed species, temperature, and concentration. Recommended experimental ranges, NO_x species, and potentially coasorbing species are provided in Gap IDs #1 and #3.	Calculation from chemical data bases, laboratory studies.	Equipment size	Concentrations of other gas species in the DOG may be low enough to limit impact of co-absorbed species on NO_x scrubbing.
7	**Selectivity** $(X_a/Y_a)/(X_b/Y_b)$ (unitless) *Where X_a and X_b are mol fractions of species a and b respectively in the adsorbed phase, and Y_a and Y_b are mol fractions of species a and b in the bulk phase*	Determination of amounts of NO_x and contaminants that adsorb (derived from Gap IDs #3–6).	Determination from chemical and engineering literature; laboratory and pilot tests.	Equipment size/operational sequencing./process safety.	Detailed models have been developed for NO_x scrubbing. Lab tests or pilot tests are needed to confirm chemical and engineering literature.

Table 6. Data gaps for NO_x absorber evaluations (continued)

#	Property *Units*	Specific requirements	Approach	Data gap impact	Notes
8	**Sorbent density** *kg/m³ absorbent liquid*	Density of aqueous solution.	Direct measurement or from manufacturer.	Equipment size	Methods exist to estimate aqueous solution density to high precision.
9	**Sorbent bulk density** *kg/m³ absorbent liquid*	N/A, liquid is a continuous phase.	N/A	N/A	
10	**Specific heat capacity** *J/K/kg*	Specific heat capacity of fresh and loaded absorbent from −15°C–50°C.	Direct measurement of fresh sorbent and loaded sorbent.	Heat duty	Estimated from properties of water and acid.
11	**Thermal conductivity** *W/m/K*	Thermal conductivity of absorbent from −15°C–50°C.	Direct measurement of fresh sorbent and loaded sorbent.	Heat duty/operational sequencing	Estimated from properties of water and acid.
12	**Radiation stability** *% degradation in capacity and/or absorption rate over time as a function of radiation exposure*	Change in scrubber DF and absorber capacity as a function of absorbed dose.	Gamma irradiation of selected scrubber solution compositions; measurement of amounts of irradiated species and reaction products.	Radiolysis producing simpler reactive molecules or causing polymerization of co-adsorbed species	Effect on neat scrubbing solution is of little concern, but how complex or reactive species form is unknown.
13	**Mechanical stability** *N/mm (load vs particle diameter)* *µg/m³ loss to gas stream*	N/A	N/A	N/A	
14	**Thermal stability** *% degradation in capacity over time at selected operating temperature*	N/A	N/A	N/A	

Table 6. Data gaps for NO_x absorber evaluations (continued)

#	Property *Units*	Specific requirements	Approach	Data gap impact	Notes
15	**Chemical stability** % degradation in capacity over time as a function of other species present in gas stream	N/A	N/A	N/A	
16	**Reactivity** *Compatibility as determined by standardized compatibility tables*	Confirmation that any compatibility issues can be avoided through selection of materials of construction, appropriate pretreatment of gas stream, operational envelope, etc.	Direct evaluation	Material life cycle	
17	**Regeneration** No. of cycles before degrading to 80% of capacity for the target element	N/A	N/A	N/A	Acid solution is recycled, main concern is accumulation of organics in the recycled acid.
18	**Desorption rate of NO_x during post loading purge** *mol/m^3 absorbent liquid /h*	N/A	N/A	N/A	This metric/gap not considered for scrubbers.
19	**Desorption of co-adsorbed species** *mol co-adsorbed species retained/m^3 sorbent*	N/A	N/A	N/A	This metric/gap not considered for scrubbers.

Table 6. Data gaps for NO_x absorber evaluations (continued)

#	Property *Units*	Specific requirements	Approach	Data gap impact	Notes
20	**Desorption rate of co-adsorbed species** *mol/m^3 absorbent liquid/h*	N/A	N/A	N/A	This metric/gap not considered for scrubbers.
21	**Purity of desorbed NO_x streams** *ppm or % NO_x*	N/A	N/A	N/A	This metric/gap not considered for scrubbers.
22	**Cooling time** *h*	N/A	N/A	N/A	This metric/gap not considered for scrubbers.
Metrics for system design and performance criterion					
23	**Pressure drop** *Pa/m vs m^2 column*	Column size required for <2.5 kPa pressure drop based on expected gas flow rates.	Direct measurement or scaled derivation.	Operational sequencing/equipment size	
24	**Decontamination factor (DF)** *$[NO_x]_{inlet}/[NO_x]_{outlet}$* (unitless)	DF data as function of sorbent, iodine species, operating temp, concentration, time, and bed depth.	Calculation/engineering design	Equipment size/operational sequencing	Overall DF depends on equivalent number of theoretical stages and solubility equilibria. Equilibria may be shifted for favorable absorption in presence of oxygen.
25	**Height of a theoretical stage** *m*	Height of a theoretical stage as a function of gas velocity, target species concentration, operating temperature, and presence of co-absorbed species shown to have impact on total capacity or absorption rate >10%.	Calculation/engineering design	Equipment size	

Table 6. Data gaps for NO_x absorber evaluations (continued)

#	Property *Units*	Specific requirements	Approach	Data gap impact	Notes
26	**Liquid volume/holdup** *m^3*	Determination of required system liquid inventory (holdup) necessary to ensure acceptable removal characteristics. Based on calculated diameter and height of tower.	Engineering design/operability choice	Equipment size	Footprint and height of scrubber to be calculated.

4.5 CO_2 recovery systems

The CO_2 recovery system follows the NO_x recovery system as shown in Figs.1 and 3. The primary gases entering CO_2 recovery include CO, CO_2, the carrier gas (usually components from air), and the noble gases. Minor components of the gas will include trace iodine, organic compounds, residual NO_x gases, and acid (HNO_3) vapors. Scrubbing of the gas stream with aqueous caustic (NaOH) solution recovers the CO_2 by a combination of sorption and chemical conversion to sodium carbonate, sodium bicarbonate, or both (Meleshyn and Noseck 2012). Depending on the fraction of carbon that is present as CO, rather than CO_2, the carbon capture performance will be degraded. The noble gases and inert components of the carrier gas are sparingly soluble in the caustic solution. Any iodine remaining in the gas stream will have the tendency to absorb into the caustic solution, but the rate will be limited by its very low concentration. Organic compounds will also be partially, or wholly, absorbed into the caustic solution, and the organic acids will be neutralized to the sodium salts of the acid. Nitric and nitrous acid vapors will be adsorbed and converted to sodium nitrate and sodium nitrite, respectively. Residual NO_x will also be converted to nitrates and nitrites, but N_2O is less reactive so a smaller fraction of it will be absorbed in the system. In the CO_2 recovery system, the carbonate-laden caustic solution is treated with calcium hydroxide [$Ca(OH)_2$] to precipitate calcium carbonate ($CaCO_3$), which can be dewatered and prepared for disposal. The disposition of the minor compounds containing iodine and organics needs to be determined. Accumulation of an organic phase may require a skimmer or other means (e.g., part of any O_2 removal system) to reduce organic vapors from entering the downstream Xe and Kr recovery system. Accumulation of organics may become a safety concern over periods of extended operation.

Like NO_x scrubbers, CO_2 scrubbers are considered relatively standard industrial equipment; however, there appears to be limited experience in the design and sizing of this equipment within the reprocessing community. Further, the experience base of staff with operating experience with remotely operated scrubbers is declining. As noted in Section 4.4, a small pilot facility to support the collection of additional design data and to train staff is needed. Key data that should be of the highest priority is the absorption capacities and impacts on waste form for both the species of interest and the residual iodine, tritium, and NO_x that may breakthrough the primary abatement systems (Table 6 Gap ID #1–7). Also of interest is the integration of a NO_x and CO_2 scrubber to determine the impacts of residual NO_x on the CO_2 scrubber performance and the subsequent waste form.

Table 7. Data gaps for CO_2 absorber evaluations

#	Property *Units*	Specific requirements	Approach	Data gap impact	Notes
Metrics for technical performance and physical and chemical characteristics criterion					
1	**Absorption Capacity** *mol/m^3 absorbent liquid*	Maximum capacity of scrub solution as a function of CO_2 concentration, temperature, and gas velocity. - Gas velocities from 0.18 to 10 m/min - Operating temperature range is −10°C–50°C - CO_2 7.1×10^{-4} kg/m^3 - CO not known	Chemical equilibrium modeling and detailed scrubber design; demonstrated in pilot-scale tests.	Equipment size	
2	**Capture removal rates for primary species** *Mol/m^3 absorbent liquid/h*	Adsorption rate data as function of CO_2 concentration, temperature, and gas velocity. Recommended experimental ranges are provided in Gap ID #1.	Chemical modeling, and measurement of CO removal with changes in reagents (e.g., H_2O_2) and operating conditions.	Equipment size	

Table 7. Data gaps for CO_2 absorber evaluations (continued)

#	Property *Units*	Specific requirements	Approach	Data gap impact	Notes
3	**Capacity for other species present in gas stream** *mol/m³ absorbent liquid*	Maximum capacity for potential co-absorbed species as a function of species, their concentrations, temperature, and gas velocity. Co-adsorbing species may include NO_2, NO, I_2, and organic acids. - Gas velocities of 0.18 to 10 m/min - Operating temperature range is −10°C–50°C - CO_2 7.1×10^{-4} kg/m³ - CO not known - I_2 7×10^{-8} kg/m³ - NO 4.3×10^{-5} kg/m³ - NO_2 1.6×10^{-4} kg/m³ - Organics not known	Column scrubber tests and/or column engineering models. Tests with steam stripping, H_2O_2 addition or other techniques.	Equipment size/operational sequencing	Escape of less easily scrubbed NO from NO_x scrubber; organic compounds, esp. organic acids.
4	**Capture rate for co-absorbed species** *Mol/m³ absorbent liquid/h*	Adsorption rate data for selected sorbent as function of co-absorbed species, temperature, and concentration. Recommended experimental ranges and potentially co-adsorbing species are provided in Gap ID #3.	Chemical modeling and solubility measurements as required.	Equipment size/operational sequencing	Acid, acid gases and iodine likely mass transfer limited—engineering design.

Table 7. Data gaps for CO_2 absorber evaluations (continued)

#	**Property** ***Units***	**Specific requirements**	**Approach**	**Data gap impact**	**Notes**
5	**Change in sorbent capacity for CO_2 in presence of other species present in gas stream** *mol/m^3 absorbent liquid*	CO_2 capacity accounting for potential adverse effects from co-absorbed species as a function of species, temperature, and concentration. Recommended experimental ranges and potentially coasorbing species are provided in Gap ID #3.	Chemical modeling and solubility measurements as required.	Equipment size	Acid, acid gases and iodine likely mass transfer limited—engineering design.
6	**Change in CO_2 capture rate in presence of co-absorbed species** *Mol/m^3 absorbent liquid/h*	CO_2 adsorption rate data for selected sorbent as function of co-absorbed species, temperature, and concentration. Recommended experimental ranges and potentially co-adsorbing species are provided in Gap ID #3.	Chemical modeling and solubility measurements as required.	Equipment size	Acid, acid gases and iodine likely mass transfer limited—engineering design.
7	**Selectivity** $(X_a/Y_a)/(X_b/Y_b)$ (unitless) *Where X_a and X_b are mol fractions of species a and b respectively in the adsorbed phase, and Y_a and Y_b are mol fractions of species a and b in the bulk phase*	Determination of amounts of CO_2 and contaminants that adsorb (derived from Gap IDs #3–6).	Chemical and engineering literature. Laboratory tests and pilot tests.	Equipment size/operational sequencing/ process safety	Engineering design methods adequate for primary function. Effect of long-term buildup of minor compounds unknown.

Table 7. Data gaps for CO_2 absorber evaluations (continued)

#	Property *Units*	Specific requirements	Approach	Data gap impact	Notes
8	**Sorbent density** *kg/m^3*	Density of aqueous solution.	Direct measurement or from manufacturer.	Equipment size	Methods exist to estimate aqueous solution density to high precision.
9	**Sorbent bulk density** *kg/m^3*	N/A, liquid is a continuous phase unless/until buildup of organics lead to a second liquid phase.	N/A	Equipment size	
10	**Specific heat capacity** *J/K/kg*	Specific heat capacity of fresh and loaded absorbent over range of operating conditions. Operating temperature range is −15°C–50°C.	Direct measurement or calculation of fresh sorbent and loaded sorbent.	Heat duty	Estimated from properties of water and soluble salts.
11	**Thermal conductivity** *W/m/K*	Thermal conductivity of absorbent over range of operating conditions. Operating temperature range is −15°C–50°C.	Calculation with direct measurement of fresh sorbent and loaded sorbent for verification, if desired.	Heat duty/operational sequencing	Estimated from properties of water and soluble salts.
12	**Radiation stability** *% degradation in capacity and/or absorption rate over time as a function of radiation exposure*	Change in scrubber DF and absorber capacity as a function of absorbed dose.	Gamma irradiation of selected scrubber solution compositions; measurement of amounts of irradiated species and reaction products, if any, along with intermittent evaluation of adsorption capacity, adsorption rate, changes in co-adsorption rates and capacities.	Radiolysis producing simpler reactive molecules or causing polymerization of co-adsorbed species	Likely little effect on scrubbing solution, but accumulation of troublesome (esp organic) compounds is unknown.

Table 7. Data gaps for CO_2 absorber evaluations (continued)

#	Property *Units*	Specific requirements	Approach	Data gap impact	Notes
13	**Mechanical stability** *N/mm (load vs particle diameter)* *$\mu g/m^3$ loss to gas stream*	N/A	N/A	N/A	
14	**Thermal stability** *% degradation in capacity over time at selected operating temperature*	N/A	N/A	N/A	
15	**Chemical stability** % degradation in capacity over time as a function of other species present in gas stream	N/A	N/A	N/A	
16	**Reactivity** *Compatibility as determined by standardized compatibility tables*	Confirmation that any compatibility issues can be avoided through selection of materials of construction, appropriate pretreatment of gas stream, operational envelope, etc.	Direct evaluation	Equipment life	
17	**Regeneration** No. of cycles before degrading to 80% of capacity for the target element	N/A	N/A	N/A	Caustic is added as needed to maintain required concentration, and some take-off is needed to expel excess water from time to time. Accumulation of organics is the main concern.

Table 7. Data gaps for CO_2 absorber evaluations (continued)

#	**Property** ***Units***	**Specific requirements**	**Approach**	**Data gap impact**	**Notes**
18	**Desorption rate of CO_2 during post loading purge** *mol/m^3 absorbent liquid /h*	N/A	N/A	N/A	This metric/gap not considered for scrubbers.
19	**Desorption of co-adsorbed species** *mol co-adsorbed species retained/ m^3 absorbent liquid*	N/A	N/A	N/A	This metric/gap not considered for scrubbers.
20	**Desorption rate of co-adsorbed species** *mol/m^3 absorbent liquid/h*	N/A	N/A	N/A	This metric/gap not considered for scrubbers.
21	**Purity of desorbed CO_2 streams** *ppm or % CO_2*	N/A	N/A	N/A	This metric/gap not considered for scrubbers.
22	**Cooling time** *h*	N/A	N/A	N/A	This metric/gap not considered for scrubbers.
Metrics for system design and performance criterion					
23	**Pressure drop** *Pa/m vs m^2 column*	Size columns and sorbent particle size for <2.5 kPa pressure drop based on expected gas flow rates.	Direct measurement or scaled derivation.	Operational sequencing/ equipment size	

Table 7. Data gaps for CO_2 absorber evaluations (continued)

#	Property *Units*	Specific requirements	Approach	Data gap impact	Notes
24	**Decontamination factor (DF)** $[CO_2]_{inlet}/[CO_2]_{outlet}$ (unitless)	Overall DF dependent on equivalent number of theoretical stages and solubility equilibria. Shifts in equilibria may be effected by addition of H_2O_2. Gas velocities of 0.18 to 10 m/min, determined by design. Operating temperature range is −10°C–50°C	Calculation/engineering design.	Equipment size/operational sequencing	
25	**Height of a theoretical stage** *m*	Height of a theoretical stage as a function of gas velocity, target species concentration, operating temperature, and presence of co-absorbed species shown to have impact on total capacity or absorption rate >10%. Same gas velocities and temperatures as Gap ID #25.	Calculation/engineering design.	Equipment size	
26	**Liquid volume/holdup** m^3	Determination of required system liquid inventory (holdup) necessary to ensure acceptable removal characteristics. Based on calculated diameter and height of tower.	Engineering design/operability choice.	Equipment size	Footprint and height of scrubber to be calculated.

4.6 Kr/Xe recovery systems

The Engineering Evaluation identified a lack of pertinent engineering data, such as the adsorption properties, desorption properties, effects of co-adsorbed species and sorbent stability over multiple cycles on solid sorbents for the removal of xenon and krypton. These data are critical to accurately design capture systems using solid sorbent materials for separation of xenon and krypton.

Table 7 summarizes the data gaps and requirements for the Xe/Kr solid phase separation system. These gaps follow the same structure as described in the criteria report but focus primarily on the properties of the sorbent materials and key operational parameters.

The most important data gap to be filled for the Xe/Kr system is to obtain data on the desorption properties of the Xe/Kr sorbents (Gap ID #18). The next most critical is the determination of the purity of the desorbed streams and their impact on the final waste stream compositions (Gap ID #21). Also of importance is the determination of the sorbent stabilities over multiple regenerations and exposures to radiation (Gap ID #12 and #17). Addressing these gaps will allow a significant refinement in the design of the Xe/Kr capture system.

The second tier data gaps include the determination of the adsorption properties (Gap ID #1–6) as these will aid in the determination of the required length of the bed to achieve the desired Xe/Kr DF.

Because of the nature of the Kr/Xe adsorption systems, deep-bed testing is required to acquire the desired data. It is envisioned many of these gaps will be investigated using the multicolumn deep-bed testing system. As the technology advances, integration with other unit operations or the ability to produce complex feed streams will be required.

The thermal properties, mechanical stability, and chemical stability data gaps are considered a lower priority at this time.

Table 8. Data gaps for Kr/Xe sorbent evaluations

#	Property *Unit*	Specific requirements	Approach	Data gap impact	Notes
Metrics for technical performance and physical and chemical characteristics criterion					
1	**Capacity** *mmol/kg sorbent*	Maximum adsorption capacity and initial breakthrough time for Kr/Xe on selected sorbent material as a function of Kr and Xe concentration, operating temperature, flow rate, and pressure. - Kr concentration range of interest is 0.004%–100% - Xe concentration of interest is 0.04%–100% - Temperature range is −80°C–37°C - Gas velocities of 0.18–10 m/min. - Pressure swing ranges requires optimization	Deep-bed tests using selected sorbent. Both temperature and pressure swing operations are required.	Equipment size	- Kr data gathered for HZ-PAN up to 4% in He, up to 0.015% in air - Xe data gathered for HZ-PAN up to 0.5% in He, up to 0.15% in air - Kr data gathered for Ag-PAN up to 4% in He, up to 0.015% in air - Xe data gathered for Ag-PAN up to 0.5% in He, up to 0.15% in air - Data gathered for MOF over limited experimental ranges
2	**Capture/ rate for primary species** *mmol/kg sorbent/h*	Adsorption rate data for selected sorbent as function of, Kr and Xe concentration operating temperature, flow rate and pressure. Recommended experimental ranges are provided in Gap ID #1.	Deep-bed tests using selected sorbent. Both temperature and pressure swing operations are required.	Equipment size	Data may be obtained from existing deep-bed testing of HZ-PAN, AgZ-PAN. Further deep-bed testing required for MOFs.

Table 8. Data gaps for Kr/Xe sorbent evaluations (continued)

#	Property *Unit*	Specific requirements	Approach	Data gap impact	Notes
3	**Capacity for other species present in gas stream** *mmol/kg sorbent*	Maximum adsorption capacity and initial breakthrough for potential co-absorbed species by selected sorbent material as a function of species, operating temp, flow rate and concentration. - Temperature range is −87°C–37°C - Gas velocities of 0.18 to 10 m/min - Pressure swing ranges requires optimization - Species of interest are N_2, O_2, Ar, H_2O, CO_2, I; - H_2O with a dew point of −6°C–90°C - O_2 concentration of up to 20% - N_2 concentration of up to 80% - CO_2 concentrations of up to 0.044% - I concentrations of up to 0.1 kg/m^3	Deep-bed tests using selected sorbent. Both temperature and pressure swing operations.	Equipment size/operational sequencing	Data for a mixture of N_2/O_2 is available from deep-bed testing of HZ-PAN, AgZ-PAN. Limited H_2O data are available. Thin-bed test data are available for MOF for N_2, O_2, H_2O, and CO_2. The combination of N_2 and O_2 are of primary concern due to overwhelming amounts. It may not be necessary to test for each individually. At minimum, H_2O should be included in future sorbent testing.

Table 8. Data gaps for Kr/Xe sorbent evaluations (continued)

#	Property *Unit*	Specific requirements	Approach	Data gap impact	Notes
4	**Capture rate for co-absorbed species** *mmol/kg sorbent /h*	Adsorption rate data for selected sorbent as function of co-absorbed species concentration, operating temp, flow rate and concentration. Recommended experimental ranges and species of interest are provided in Gap ID #3.	Deep-bed tests using selected sorbent.	Equipment size/operational sequencing	Same as #3
5	**Change in sorbent capacity for Kr, Xe in presence of other species present in gas stream** *mmol/kg sorbent*	Kr/Xe capacity of selected sorbent material in the presence of co-absorbed species as a function of species, operating temp, flow rate and concentration. Recommended experimental ranges and species of interest are provided in Gap ID #3.	Deep-bed tests using selected sorbents.	Equipment size	Same as #3.
6	**Change in Kr, Xe capture rate in presence of co-absorbed species** *mmol/kg sorbent /h*	Kr/Xe adsorption rate data for selected sorbent as function of co-absorbed species, operating temp, and concentration. Recommended experimental ranges and species of interest are provided in Gap ID #3.	Deep-bed tests using selected sorbents.	Equipment size	Data gathered during cycling and regeneration tests.
7	**Selectivity** $(X_a/Y_a)/(X_b/Y_b)$ (unitless)	Derived from data on co-absorption studies shown above.	Deep-bed tests using selected sorbent.	Equipment size/operational sequencing	

Table 8. Data gaps for Kr/Xe sorbent evaluations (continued)

#	Property *Unit*	Specific requirements	Approach	Data gap impact	Notes
8	**Sorbent particle density** kg/m^3 *sorbent*	Density of individual particles in kg/m^3.	Direct measurement.	Equipment size	Completed for HZ-PAN, AgZ-PAN, CaSDB MOF.
9	**Sorbent bulk density** *kg/m^3 sorbent*	Bulk density of bed of sorbent material in typical loading configuration in kg/m^3.	Direct measurement.	Equipment size	Completed for HZ-PAN, AgZ-PAN, CaSDB MOF. However, this will change somewhat based on sorbent particle size chosen based on column design.
10	**Specific heat capacity** *J/K/kg or J/K/m^3*	Specific heat capacity of fresh and loaded sorbent over range of operating conditions. Operating temperature range is −80°C–150°C.	Direct measurement of fresh and loaded sorbent.	Heat duty	Currently estimated for HZ-PAN, AgZ-PAN based on published data for individual components of the composite material.
11	**Thermal conductivity** *W/m/K*	Thermal conductivity of sorbent over range of operating conditions. Operating temperature range is −80°C–150°C.	Direct measurement of fresh and loaded sorbent.	Heat duty/operational sequencing	Currently estimated for HZ-PAN, AgZ-PAN based on published data for individual components of the composite material.
12	**Radiation stability** *Δ mmol/kg sorbent/kGy*	% degradation in capacity and/or adsorption rate over time as a function of radiation exposure. Total cumulative adsorbed dose for Kr, Xe sorbents is design dependent.	Gamma and/or beta irradiation of selected adsorbent to doses TBD, along with intermittent evaluation of adsorption capacity, adsorption rate, changes in co-adsorption rates and capacities.	Material life cycle	Testing has indicated that a high radiation dose may have an impact on HZPAN and/or AgZPAN sorbent performance, Further evaluations will be needed to define what dose is damaging.

Table 8. Data gaps for Kr/Xe sorbent evaluations (continued)

#	**Property** ***Unit***	**Specific requirements**	**Approach**	**Data gap impact**	**Notes**
13	**Mechanical stability** *N/mm (load vs particle diameter)* *µg/kg sorbent loss to gas stream*	Determination of the fines generation and mechanical strength of the sorbent material.	Direct measurement. *Measurements of both particle crush strength and abrasion resistance will be required.*	Material life cycle	Testing of particle crush strength of HZ-PAN, AgZ-PAN and CaSDB MOF is complete and deemed sufficient, but not published. One test of abrasion resistance for HZ-PAN and AgZ-PAN is complete with no generated fines. Packed column crush strength has not been measured. One test of abrasion resistance for CaSDB MOF is complete with indication of excessive fines generation. Further testing/engineered form modification should be considered. Packed column crush strength testing has not been done on any sorbent to date.
14	**Thermal stability** *% degradation in capacity over time at selected operating temperature*	Determine the extent of capacity loss due to extended exposure to operating temperature. Determine the extent of capacity loss due to thermal cycling and excursions.	Data may be gathered from existing long-term testing of selected sorbents through multiple adsorption/desorption cycles.	Material life cycle	

Table 8. Data gaps for Kr/Xe sorbent evaluations (continued)

#	Property *Unit*	Specific requirements	Approach	Data gap impact	Notes
15	**Chemical stability** *% degradation in capacity over time as a function of other species present in gas stream*	Determine the extent of capacity loss due to extended exposure to other species present in the off-gas stream.	Extended testing. *Age sorbents for various lengths of time to elevated levels of each of the non-target species expected. Determine Kr/Xe capacity and rates of adsorption. Differences should be attributable to aging and should be a function of exposure time and non-target species exposure.*	Material life cycle	AgZ-PAN, HZ-PAN, and MOF materials tested to date meet this criterion.
16	**Reactivity** *Compatibility as determined by standardized compatibility tables*	Confirmation that any compatibility issues can be avoided through selection of materials of construction, appropriate pretreatment of gas stream, operational envelope, etc.	Direct evaluation.	Material life cycle	AgZ-PAN, HZ-PAN, and MOF materials tested to date meet this criterion.
17	**Regeneration capacity stability** *Δ mmol/kg sorbent /regeneration cycle*	No. of cycles before degrading to 80% of capacity for the target element	Long-term, repeated testing of select sorbent over multiple adsorption/desorption cycles.	Material life cycle	# of thermal cycles have been reported for AgZ-PAN under defined temp ranges.
18	**Desorption rate of Kr/Xe** *mmol/kg sorbent/h*	Desorption of Kr and Xe as a function of operating temperature, purge gas flow rate and pressure. Recommended experimental ranges are provided in Gap ID #1	Deep-bed tests using selected sorbent. Both temperature and pressure swing operations.	Operational sequencing	Some data are available for existing sorbents.

Table 8. Data gaps for Kr/Xe sorbent evaluations (continued)

#	Property *Unit*	Specific requirements	Approach	Data gap impact	Notes
19	**Desorption of co-adsorbed species** *mmol retained/kg sorbent*	% of co-adsorbed species desorbed during selected regeneration conditions. Recommended experimental ranges and potentially co-adsorbing species are provided in Gap ID #3.	Deep-bed tests using selected sorbent.	Equipment size	Some data may be obtained from existing deep bed testing of HZ-PAN, AgZ-PAN, but further testing is required. Further deep bed testing is required for MOFs.
20	**Desorption rate of co-adsorbed species** *mmol/kg sorbent/h*	Desorption rate as a function of desorption temperature and purge stream gas velocity. Recommended experimental ranges and potentially co-adsorbing species are provided in Gap ID #3.	Deep-bed tests using selected sorbent collecting and analyzing the effluent from the sorbent bed.	Material life cycle	Some data may be obtained from existing deep bed testing of HZ-PAN, AgZ-PAN, but further testing is required. Further deep-bed testing is required for MOFs.
21	**Purity of desorbed Xe/Kr streams** *ppm or % Xe/Kr*	Concentrations of the captured gases desorbed from the sorbent after loading. Determination of Kr and Xe concentration factors and purity.	Deep-bed tests using selected sorbent.	Operational sequencing	Preliminary testing has shown this composition is somewhat difficult to accurately define.
22	**Cooling time** *h*	Hours to cool from regeneration temperatures to adsorbing temperatures.	Derived from heat capacity, thermal conductivity, and bed design.	Heat duty	Readily derived and confirmed during deep-bed testing.
Metrics for system design and performance criterion					
23	**Pressure drop** *kPa/m vs m^2 column*	Size columns and sorbent particle size for <2.5 kPa pressure drop based on expected gas flow rates	Direct measurement or scaled derivation.	Operational sequencing/ equipment size	

Table 8. Data gaps for Kr/Xe sorbent evaluations (continued)

#	**Property** *Unit*	Specific requirements	Approach	Data gap impact	Notes
24	**Decontamination factor (DF)** *[Isotope]$_a$/ [Isotope]$_b$* (unitless)	DF data as function of sorbent, operating temperature, Kr/Xe concentration, time, and bed depth. Recommended experimental ranges are provided in Gap ID #1.	Deep-bed testing with online measurement of inlet and effluent H_2O concentrations. *Test system must be able to hold a deep bed of sorbent at given temperature and expose sorbent to a generated air stream containing water (and any potentially co-adsorbed species) at varying concentrations within the specified concentration range and gas velocity. Both inlet and effluent gas streams must be assessed for Kr/Xe concentration.*	Equipment size/operational sequencing	This is actually a function of the overall system design but the ability to demonstrate that the assumed DF is achievable is needed for a comparable design.
25	**Length of Mass Transfer Zone (MTZ)** *m*	Length of MTZ as a function of gas velocity, target species concentration, operating temperature, and presence of co-absorbed species shown to have impact on total capacity or adsorption rate >10%. Recommended experimental ranges are provided in Gap ID #1.	Deep-bed tests with bed depth that captures 1.5 times MTZ. Predictions can be made from thin-bed tests and data may also be determined from DF testing using deep beds. *(Deep-bed testing: Gap ID #24).*	Equipment size	Derived from deep-bed test breakthrough curves.

5. CONCLUSIONS AND RECOMMENDATIONS

5.1 Summary of priority data gaps

In the development of this report considerable discussion occurred on the lack of data regarding the source terms. During the Engineering Evaluation conducted in FY 2016, the design of a Ru capture system for the TPT system was hindered because of the lack of critical data on Ru generation and speciation. Also, the volatilization of other semi-volatile components from the TPT system was ignored. Better understanding and quantification of the off-gas streams from the TPT—and to a lesser degree the dissolution system—is critical. However critical, such work lies somewhat outside the current scope of the Off-gas Sigma Team. That said, joint studies into this critical interface area should be undertaken at the earliest opportunity.

The Engineering Evaluation pointed out that although considerable progress has been made in developing abatement systems for the four primary volatile radionuclides of interest (i.e., ^{3}H, ^{14}C, ^{85}Kr, and ^{129}I) much of the data focused on the capacity of the adsorbent or absorbent for the species of interest only and little or no data existed for the co-absorbed species.

For the Ru sorbents, the most critical gap beyond that of the source term is adsorption capacity and adsorption rate data. Because this may be one of the first adsorption processes in the off-gas train, the co-adsorption of other materials in the off-gas stream is also a high priority. The next most important gap to address is the determination of the MTZ and its length.

For tritium, the most substantial data gaps that limit the refinement of the tritium removal system design are related to the co-adsorption/desorption of other off-gas species. Since this is envisioned to be a regenerable system, testing of a multicolumn configuration of the capture and regeneration systems coupled with the tritium recovery, concentration, and solidification processes should be demonstrated in an integrated fashion.

The iodine gaps were divided into gaps for DOG systems and for VOG/air TPT systems. For the DOG system, data on the impact of co-adsorbed species and the length of the MTZ are judged as the most pressing need. Thin- and deep-bed test systems similar to the current systems are required to collect these data. Data on the desorption behavior of co-absorbed species as well as target species should also be a near term priority and should be collected as part of the adsorption testing. For VOG, and air TPTOG systems additional basic adsorption performance data are still needed for design of these iodine sorption systems. For these systems, current R&D efforts are focused mainly on the adsorption capacity using deep beds for very dilute iodine streams. Because of the time involved in collecting this data, additional tests systems should be established. Obtaining data on the expected efficiency of the iodine sorbents under VOG and air TPT conditions in terms of DF and length of the associated MTZ are also key data needs. Closure of these data gaps requires test systems designed to operate with deep beds and over extended test durations (months to years).

There are fewer data gaps for scrubbers than for the other systems. The Engineering Evaluation drew from existing designs and industrial experience. What may become a gap is the limited experience within the Department of Energy complex to design such systems as very little work has been conducted in this area for two to three decades. The impacts of co-absorbed species, especially from NO_x and iodine that pass through upstream operations, on the scrubber design and operation and on the waste form also need to be better evaluated.

The most important data gap to be filled for the Xe/Kr system is to obtain data on the desorption properties of the Xe/Kr sorbents. The next most critical is the determination of the purity of the desorbed streams and their impact on the final waste stream compositions. Also of importance is the determination of the sorbent stabilities over multiple regenerations and exposures to radiation. Addressing these gaps will allow a significant refinement in the design of the Xe/Kr capture system.

5.2 Common elements

5.2.1 Capacity and adsorption rate

Capacity and adsorption rate data needs appear common for all the unit operations considered. These two parameters dictate the dimensions and operating conditions of the sorbent beds and wet scrubber systems. The capacity dictates the mass and volume of Ru, I, and Kr/Xe sorbent beds for a given adsorption duration; the adsorption rate dictates the needed depth of the solid sorbent beds and the height of the wet scrubbers. The capacity of the solid sorbents and the scrubber solutions also dictates the mass and volume of spent sorbents and scrub solutions for recycle or disposal. The maturity and quality of the engineering designs and process operating conditions are only as good as the quality of these data for the specific applications.

Continued R&D as summarized in this document is needed to mature the capture technologies as well as design and operate capture systems that use these technologies. The needed R&D generally includes laboratory and small pilot-scale thin-bed and deep-bed solid sorbent testing to expand the range of tests completed to date; and eventually scaled up to integrated systems using representative synthetically generated gas mixtures or slip-streams off of operating reprocessing facilities.

5.2.2 Co-adsorption of non-target species

Co-adsorption of other species is important to the extent that it interferes with the adsorption of the target species, impacts process operation, or impacts handling, recycle, or disposal of the spent sorbent/scrub solutions, and the waste form and disposal of the captured species. This is of secondary importance compared to demonstrating and optimizing capacity and adsorption rates for the target species; but co-adsorption and impacts of co-adsorption are areas where generally even less is currently known for both sorbent beds and wet scrubber systems. Co-adsorption data and impacts of co-adsorption on the adsorption and waste form/disposal processes should be obtained in parallel with maturing the adsorption technologies for the target species. Desorption tests may require larger, more complex test beds to provide suitable simulated feed streams and the subsequent desorption, effluent sampling, and sample analysis of the associated co-adsorbed species.

5.2.3 Sorbent physical properties data

Sorbent particle and bulk densities impact the volume of the adsorption system for a given mass of sorbent. Although these are important parameters for sizing sorbent bed systems, obtaining these data does not require extensive test programs.

5.2.4 Regenerable systems

For regenerable systems, data on desorption and multicycle operations is also critical for the sizing of the subsequent recovery systems, the design of any additional purification, and the conversion to a waste form. In general, obtaining this type of data requires larger, more complex test beds and must deal with co-adsorbed species, which are needed for regenerable sorbents like those being tested for Kr and Xe capture, and scrub solution for CO_2 that is recycled back to the scrubber after precipitating out the captured CO_2. Considerable uncertainty exists in how to engineer regeneration systems for Kr and Xe capture, considering (1) expected height and footprint limitations, and (2) how to implement regeneration using either pressure, temperature, or combined pressure-temperature swing systems. Regenerable sorbents and sorbent beds must be able to operate in the process for potentially orders of magnitude longer time than non-regenerable sorbents. The design and operation of thermal swing regeneration systems must rely on heat transfer constraints imposed by sorbent heat capacities and thermal conductivities and account for thermal, mechanical, chemical, and radiation stabilities over the design temperature ranges. Mechanical, chemical, and radiation stabilities also apply, along with designs for pressure and radiological control, to pressure swing systems.

The general maturity level of wet scrubber systems is high. NO_x and CO_2 scrubber systems can be designed using standard wet scrubber engineering design, with adaptations from references cited in this report. However, the actual use of these technologies in aqueous reprocessing dissolver off-gas applications has been limited in the United States. Uncertainties in actual process efficiencies, impacts of co-adsorbed species, and the properties and amounts of the resultant nitric acid recycle and carbonate disposal streams will need to be resolved to mature these technologies for these specific applications.

5.3 Integrated testing

Many of the data gaps identified in this report can be effectively addressed with test systems focused on specific target component. In the case of regenerable sorbents that focus on tritium, Xe, and Kr, more complex test beds will be required to obtain data on the desorption behavior, co-absorbed species, potential product purification, and cycle time systems. In the case of Xe and Kr, some preconditioning of the gas streams may also be required in an actual plant design. These systems should be integrated with the Xe/Kr capture systems.

Integrated test beds should also be designed that couple the tritium capture and recovery systems and iodine capture systems with the Ru abatement system. This should be completed for both the TPTOG stream conditions as well as the dissolver off-gas system conditions, with a focus on the long-term effects of water vapor on the Ru recovery system and the potential accumulation of Ru within the tritium capture systems. Of importance is the evaluation of the long-term effects of water vapor on the Ru sorbent, the contribution of the back-flushable filters and HEPA filters to the overall Ru DF, and the selective recovery of tritium-contaminated water for conversion to a stable waste form.

Ultimately a fully integrated cold or tracer level off-gas test would be needed to validate any design for a future reprocessing plant. Before the design of such a system many of the data gaps identified in this report must be closed.

6. REFERENCES

Bruffey, S. H., B. B. Spencer, D. M. Strachan, R. T. Jubin, N. Soelberg, and B. J. Riley, 2015, "A Literature Survey to Identify Potentially Problematic Volatile Iodine-Bearing Species Present in Off-Gas Streams," FCR&D-MRWFD-2015-000421, ORNL-SPR-2015/290, INL/EXT-15-35609, 30 June.

Counce, R. M. and Groenier, W. S., 1978. *Nitrogen Oxide Absorption into Water and Dilute Nitric Acid—Description of a Mathematical Model and Preliminary Scrubbing Results Using an Engineering-Scale Sieve-Plate Column*, ORNL/TM-5910, Oak Ridge National Laboratory, July 1978. (Applied Technology).

Counce, R. M.,1978. *Nitrogen Oxide Absorption into Water and Dilute Nitric Acid in an Engineering-Scale Sieve-Plate Column—Description of a Mathematical Model and Comparison with Experimental Data*, ORNL-5438, Oak Ridge National Laboratory, September 1978. (Applied Technology).

Counce, R. M. and Perona, J. J.,1980. "A Mathematical Model for Nitrogen Oxide Absorption in a Sieve-Plate Column," *I&EC Process Design and Development*, vol 19, pp 426–431, July 1980.

Counce, R. M. and Perona, J. J.,1979a. "Gas-Liquid Interfacial Area of a Sieve-Plate with Downcomers and 0.6% Perforation," *I&EC Process Design and Development*, vol 18, pp 562–564, July 1979.

Counce, R. M. and Perona, J. J.,1979b. "Gaseous Nitrogen Oxide Absorption in a Sieve-Plate Column" *I&EC Fundamentals*, vol 18, no. 4, pp 400–406, November 1979.

Counce, R. M., 1979. *A User's Manual to ABNOX, a Computer Program Designed to Simulate the Nitrogen Oxide Removal Efficiency of a Multistage Bubble-Cap Tower*, ORNL/TM-6938, Oak Ridge National Laboratory, September 1979.

Counce, R. M., 1980. *The Scrubbing of Gaseous Nitrogen Oxides in Packed Towers*, ORNL-5676, Oak Ridge National Laboratory, November 1980.

Jubin, R. T., D. W. Ramey, R. S. Owens, J. M. Giaquinto, E. S. Meyers, B. B. Spencer, and G. D. Del Cul, (2009) "Voloxidation Off-Gas Capture: Experimental Results from Processing of CETE Run 2 Fuels, Report No. ORNL/TM-2009/187. Oak Ridge National Laboratory, Oak Ridge, TN. (DRAFT).

Jubin, R. T., Jordan, J. A., Spencer, B. B., Soelberg, N. R., Welty, A. K., Greenhalgh, M., Strachan, D. M., Thallapally, P. K. 2016a. *Engineering Evaluation of an Integrated Off-Gas Treatment System for Used Nuclear Fuel Reprocessing Facilities*. Report No. FCRD-MRWFD-2016-000313, Oak Ridge, TN: UT-Battelle, LLC, Oak Ridge National Laboratory.

Jubin, R. T., S. H. Bruffey, D. M. Strachan, N. R. Soelberg, and B. B. Spencer, and B. J. Riley, 2016b, "Performance Criteria for Capture and/or Immobilization Technologies," FCRD-MRWFD-2016-000533, ORNL/TM/2016-73, February, 2016.

Jubin, R. T., D. M. Strachan, and N.R. Soelberg, 2013, "Iodine Pathways and Off-Gas Stream Characteristics Characteristics for Aqueous Reprocessing Plants – A Literature Survey and Assessment," FCRD-SWF-2013-000308, ORNL/LTR-2013/383, INL/EXT-13-30119, September 15.

Jubin, R. T., N. R. Soelberg, D. M. Strachan, and G. Ilas. 2012. *Fuel Age Impacts on Gaseous Fission Product Capture during Separations.* Report No. FCRD-SWF-2012-000089, Oak Ridge, TN: UT-Battelle, LLC, Oak Ridge National Laboratory.

Jubin, R. T. and B. B. Spencer. 2017. *Evaluation of Tritium Management Approaches on Tritium Waste Volumes in Reprocessing Plants.* Report No. NTRD-MRWFD-2017-000267 and ORNL/TM-2017/307, Oak Ridge, TN: UT-Battelle, LLC, Oak Ridge National Laboratory.

Law, J., N. Soelberg, T. Todd, G. Fredrickson, S. Frank, (INL), C. Pereira, M. Williamson, W. Ebert, J. Willit (ANL), R. Jubin, (ORNL), J. Vienna, G. Lumetta, J. Crum (PNNL), T. Rudisill (SRNL), J. Bresee (DOE-NE), C. Phillips, B. Willis (EnergySolutions), S. Bader (AREVA), 2015, "Separation and Waste Form Campaign Full Recycle Case Study," FCRD-MRWFD-2015-000241, September 30.

Meleshyn, A. and Noseck, U., 2012. Radionuclide Inventory of Vitrified Waste after Spent Nuclear Fuel Reprocessing at La Hague, GRS-294, Gesellschaft für Anlagen-und Reaktorsicherheit, September 2012.

Nenoff, T., M. Rodriguez, N. Soelberg, K. Chapman, 2014, "Silver-Mordenite for Radiologic Gas Capture from Complex Streams: Dual Catalytic CH3I Decomposition and I Confinement," Microporous and Mesoporous Materials, MICMAT6530, INL/JOU-14-31276, 9 May.

Soelberg, N. R., T. G. Garn, M. R. Greenhalgh, J. D. Law, R. Jubin, D. M. Strachan, and P. K. Thallapally, 2013, "Radioactive Iodine and Krypton Control for Nuclear Fuel Reprocessing Facilities," Hindawi Publishing Corporation, Science and Technology of Nuclear Installations, Volume 2013, Article ID 702496, 12 pages, INL/JOU-13-28481, Feb. 19, 2013, http://dx.doi.org/10.1155/2013/702496.

Soelberg, N., 2013, "Recent Deep-Bed Test Results for Capturing I-129 in Used Fuel Reprocessing Off-gas Streams," Global 2013, October 3.

Soelberg, N. and T. Watson, 2014, "Phase 2 Methyl Iodide Deep-Bed Adsorption Tests," FCRD-SWF-2014-000273, INL/EXT-14-33269, September 30.

Soelberg, N. and T. Watson, 2015, "FY-2015 Methyl Iodide Deep-Bed Adsorption Test Report," FCRD-MRWFD-2015-000267, INL/EXT-15-36817, Idaho National Laboratory, September 30.

Soelberg, N., 2016, "Deep-Bed Iodine Adsorption Test Results," INL/MIS-16-38237, MRWFD Campaign Working Group Meeting, PNNL, Richland, WA, March 22-23.

Soelberg, N. and T. Watson, 2016, "FY-2016 Methyl Iodide Higher NOx Adsorption Test Report," FCRD-MRWFD-2016-000352, INL/EXT-16-40087, Idaho National Laboratory, September 29.

Welty, A.K., Garn, Troy G., and Greenhalgh, Mitchell, "Superficial Velocity Effects on HZ-PAN and AgZ-PAN for Kr/Xe Capture, Idaho National Laboratory, April 1, 2016, FCRD-MRWD-2016-000024