Performance Criteria for Capture and/or Immobilization Technologies, Revision 1

Fuel Cycle Research & Development

Prepared for
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SUMMARY

The capture and subsequent immobilization of regulated volatile radionuclides from the off-gas streams of a used nuclear fuel (UNF) reprocessing facility has been a topic of substantial research interest for the US Department of Energy and its international counterparts. Removal of specific radionuclides from the plant effluent streams before discharge to the environment is required to meet regulations set forth by the US Environmental Protection Agency. Upon removal, the radionuclides, as well as associated sorbents that cannot be regenerated in a cost-effective manner, are destined for conversion to a waste form. Research in separation and capture methodologies has included a wide range of technology types, and studies of waste forms are correspondingly diverse. In considering the future development and implementation of both sorbents and waste forms, it is necessary to identify benchmark measures of performance to objectively evaluate each sorbent system or waste form.

Sets of performance criteria and associated metrics have been developed for sorbent and waste form evaluation. These criteria address physical, radiological, and chemical characteristics, technical practicality, technical maturity, cost, and, for sorbents, system performance. The criteria and metrics appear to be robust and should be applicable despite the eventual waste classification (as either high- or low-level waste). They are flexible enough to address both aqueous reprocessing and electrochemical reprocessing of UNF. These criteria sets can serve as tools to evaluate performance at multiple stages within the development process, and in this revision (Revision 1) they have been used to assess technologies relating to krypton/xenon separations and iodine capture from off-gas streams arising from UNF reprocessing.

Assessment of krypton/xenon separations using engineered forms of two zeolite minerals (silver mordenite and hydrogen mordenite in a polyacrylonitrile-based binder [AgZ-PAN/HZ-PAN]) found that the zeolite-based separation is relatively advanced in its development, but several key issues require resolution. First, desorption processes for both krypton and xenon require refinement to provide an understanding of the product purity that can be achieved. Second, adsorption rate data is needed in order to calculate the bed depth required for effective separation. Finally, it is strongly recommended that a technical review of krypton/xenon separation by AgZ-PAN/HZ-PAN be performed to synergize available data and assess the cost savings and operational benefits that may be realized from implementation of this technology.

Assessment of metal organic frameworks (MOFs) for their use in the separation of krypton/xenon found that the ideal separation would be performed using a single-column system with a MOF selective for krypton over xenon. A robust research effort should work to identify a krypton-selective MOF designed to operate at temperatures of approximately 0°C or higher, which could be preferred over cryogenic krypton/xenon capture for used fuel reprocessing off-gas streams. In the case of the CaSDB-MOF (the most well-understood xenon sorbent to date), two issues are judged of high importance. First, xenon breakthrough capacity for the CaSDB-MOF in prototypical conditions should be determined. Preliminary research indicates that breakthrough may be near immediate, presenting a substantial obstacle in separative system design. Second, development of desorption methodology should be performed to determine regeneration time, energy requirements, and the product stream composition.

Silver-based sorbents (AgZ and AgAero) for use in iodine capture from the dissolver off-gas were evaluated against the established criteria. These sorbents are significantly better understood for this application as a result of research efforts over the past decade. The potential implementation of AgAero at a large scale is hindered by its physical degradation by components of the dissolver off-gas stream.

Less is known about the adsorption of iodine by these sorbents from other off-gas streams in the plant. Initial experimental efforts have been closely coordinated in an effort to understand organic iodine (such as would be found in the vessel off-gas) adsorption by AgZ and AgAero. Future work should expand this

experimental program, and analysis of other reprocessing facility off-gas streams such as the vitrification off-gas stream should be conducted to better understand other potential applications for iodine sorbents.

A review of iodine waste form development shows that this area is diverse and that multiple promising waste forms have been identified for the immobilization of radioactive iodine. Efforts related to the direct conversion of iodine sorbents (including AgZ and AgAero) should be continued because of the advantages of direct conversion in a waste management strategy and other sorbents should continue to be advanced as merited.

CONTENTS

FIGURES

TABLES

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ACRONYMS

AgAero	silver-functionalized silica aerogel
AgZ	silver mordenite
AgZ-PAN	silver mordenite in a polyacrylonitrile-based engineered form
ANSI	American National Standards Institute
ASTM	American Society for Testing and Materials
DF	decontamination factor
DOE	US Department of Energy
DOG	dissolver off-gas
EPA	Environmental Protection Agency
HZ-PAN	hydrogen mordenite in a polyacrylonitrile-based engineered form
HEPA	high-efficiency particle air
HIP	hot isostatic pressing
INL	Idaho National Laboratory
MOF	metal organic framework
MOX	mixed oxide (UO_2–PuO_2)
NFPA	National Fire Protection Association
ORNL	Oak Ridge National Laboratory
PAN	polyacrylonitrile
PNNL	Pacific Northwest National Laboratory
SPS	spark plasma sintering
TRL	technology readiness level
UNF	used nuclear fuel
UOX	uranium oxide (UO_2)
VOG	vessel off-gas

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PERFORMANCE CRITERIA FOR CAPTURE AND/OR IMMOBILIZATION TECHNOLOGIES, REVISION 1

1. INTRODUCTION

The capture and subsequent immobilization of the four regulated volatile radionuclides (^{3}H, ^{14}C, ^{85}Kr, and ^{129}I) from the off-gas streams of an aqueous used nuclear fuel (UNF) reprocessing facility has been a topic of substantial research interest for the US Department of Energy (DOE) and its international colleagues. Removal of some or all of these radionuclides from plant effluent streams before discharge to the environment is required to meet regulations set forth by the US Environmental Protection Agency (EPA). The extent of removal will be dependent upon fuel burnup, fuel type, cooling time, and other related factors. Upon removal, the radionuclides, as well as associated sorbents that cannot be regenerated in a cost-effective manner, are destined for conversion to a waste form. Research in separation and capture methodologies has included a wide range of technologies, including liquid caustic scrubbing systems, solid adsorbents, and cryogenic distillation. The studies of waste forms have been correspondingly diverse. In considering the technologies available for future development and implementation of both sorbents and waste forms, it is necessary to identify benchmark measures of performance to objectively evaluate each sorbent system or waste form.

In 2016, the initial issuance of this document provided guidance on the types of performance metrics that should be used to evaluate off-gas capture materials and waste forms intended for use in the recycling of UNF. For capture materials, general performance measures applicable to capture technologies for all four radionuclides were identified first, followed by a discussion of any metrics that may be specific only to a select radionuclide and its capture possibilities, where appropriate. For waste forms, the performance criteria and metrics were more generically identified without any judgement in this document about waste classification as high-level waste or low-level waste, or any presumption about specific transportation, storage, or disposal site waste acceptance criteria, beyond the following acknowledgements:

- Canister and package size and weight limits should be expected.
- Free liquids, fines, chemical reactivity, thermal, and mechanical stability limits should be expected.
- All release rates shall be controlled to ensure that the dose to the public over time from the released radionuclide must be within dose limits. The actual value(s) will be developed when a repository is selected and a total system performance analysis has been performed.

The focal point of the report is then contained in two main sections addressing criteria relevant to the evaluation of the capture media/systems and waste form performance. Suggested units and desired trends are noted, along with comments regarding the significance of each metric. The original issuance of the document in 2016 was focused on the four volatile radionuclides released during aqueous reprocessing of UNF, but the criteria identified are broadly applicable and can be used to evaluate off-gas capture and immobilization technologies for use in the electrochemical processing of UNF or in other applications, such as the off-gas treatment required in some advanced reactor designs.

This revision [Revision 1] is intended to provide an update on recent research conducted by the US DOE focused on the capture and separation of krypton (including ^{85}Kr) from xenon, the capture of iodine (including ^{129}I), and the development of iodine waste forms. Technology development related to ^{3}H and ^{14}C management is not addressed. Capture and separation of krypton, capture of iodine, and the development of iodine waste forms are all active international research areas, and the body of work contributed by US DOE-launched projects is significant. The development of each technology is evaluated using the criteria developed in this report and any outstanding fundamental questions are identified.

The following technologies are included in this evaluation:

1) The separation of krypton and xenon using engineered forms of zeolite minerals [(silver mordenite and hydrogen mordenite in a polyacrylonitrile-based binder (AgZ-PAN/HZ-PAN)]
2) The separation of krypton and xenon using CaSDB, a metal organic framework (MOF)-based solid sorbent
3) The removal of iodine from off-gas streams using silver-exchanged mordenite (AgZ)
4) The removal of iodine from off-gas streams using silver-functionalized silica aerogel (AgAero)

Although not formally evaluated against the included criteria, recently studied iodine waste forms were surveyed and are discussed in Section 6.

Application of the developed criteria sets is to be used as a tool and may be deployed differently based on the objectives and specifics of the analysis. The relative importance of individual metrics for both sorbents and waste forms may differ based on the specific properties of the radionuclide that the sorbent is designed to capture, the waste form it is designed to immobilize, the type of deployment scenario envisioned, or a myriad of other factors. For example, the selectivity of a sorbent could be of high importance for ^{85}Kr, and it could result in a significant increase in waste volume should the sorbent also capture xenon. However, the chemical stability of a ^{85}Kr sorbent is of less importance than other metrics in a reprocessing scheme in which the noble gases are captured after the off-gas stream has been treated and many of the most reactive compounds have been removed. The development of the importance or "value" of each metric within each of the criteria has not been included in this document, and the user may prioritize the criteria and metrics based on their specific application.

2. REGULATIONS RELATED TO SORBENT AND WASTE FORM PERFORMANCE

The processes by which sorbent materials, separation processes, and waste forms are selected must begin with an evaluation of their ability to contribute to regulatory compliance. In the case of sorbent materials and separation processes used to remove the four regulated volatile radionuclides from a particular waste stream, the regulatory requirements, when applied to an aqueous reprocessing facility, result in a range of decontamination factor (DF) values that depend on the characteristics of the fuel being processed, the processing rate, and other considerations (Jubin et al. 2012a). For waste form materials, the physical and chemical attributes are dictated by what is needed to allow a future candidate repository to meet performance guidelines that have yet to be set by the regulator. Since there is no candidate repository and, hence, the chemical and physical properties for waste forms are largely unknown, our approach is to provide a list of physical and chemical properties that should be an important part of a viable waste form. This list was checked qualitatively against the waste form properties that are currently available for the waste that was destined for the Yucca Mountain repository (DOE 2012). The metrics listed in the tables in this document are consistent with those required for the Yucca Mountain repository, but the properties needed for a viable waste form going to Yucca Mountain are a subset of those shown in Table 4-1. Ultimately, the materials selection process is complex and involves trade-offs between material properties, cost-to-benefit studies, technology readiness assessments (DOD 2011), and so on.

3. CAPTURE MEDIA CRITERIA

Five criteria were identified for the capture material and related systems, which include (1) technical performance and characteristics (physical and chemical properties), (2) technical practicality, (3) system design and performance, (4) technical maturity, and (5) cost. In general, these apply to capture materials and systems for all four radionuclides of interest. For each of these five criteria, multiple metrics have been identified that attempt to describe the important aspects of that criterion. Table 3-2 lists the five criteria and the associated metrics. Also included in the table are the preferred reporting units and the preferred trend in a specific metric. General descriptions for each metric are also provided. In a few cases, there is no clear desired trend. Rather, the value for that metric should be evaluated in the context of the specific radionuclide properties and ultimate implications for process design.

The metrics identified in Table 3-2 tend to focus on solid sorbents that lend themselves to use in packed beds. The evaluation of the metrics for the performance of the capture system assumes a properly designed system that optimizes the relevant performance factors to achieve the desired DF.

The system design and performance criterion include three metrics: pressure drop, DF, and bed volume. Unlike the other four criteria sets in which the values tend to be intrinsic properties of the capture system, the values for these metrics will vary based upon system design. Additionally, they are interrelated, and the values contained within these criteria should be judged as a group. For example, the pressure drop over a sorbent bed can be reduced by increasing the diameter of the sorbent column and decreasing the gas velocity of the stream to be treated. Although this would likely not affect the DF, there would be a corresponding increase in bed volume. An ideal capture system should be designed to optimize all three metrics.

3.1 Discussion of Selected Metrics

In Tables 3-2 and 4-1, there are several metrics that merit a level of discussion that is not contained within the tables. These items are marked with an asterisk (*) in the tables and expanded on in further detail here.

Capacity—This metric can be applied in different ways based on the technology to be evaluated. If the adsorbent forms stable chemical compounds with the element or compound to be removed (chemisorption), then the capacity reflects a fixed saturation value that can likely be achieved under a variety of operational conditions. Adsorbents that function by physisorption (including noble gas sorbents) will be governed according to reversible isotherms, and the saturation capacity will be dependent on the partial pressure of the sorbate and the operating temperature of the system. Other types of capacity that may be substituted as needed include the breakthrough capacity of a given material or the working capacity of the material.

Mechanical Stability—This property is a measure of the ability of the sorbent material to avoid being crushed when compressed or to avoid attrition of the sorbent material and dust generation during use. Air passing through a packed column can cause vibration or movement of the particles and subsequent dust generation from fragile particles. The objective is to make sure that the material does not (a) generate a quantity of dust that could cause the bed to decrease significantly in mass as a function of time, (b) generate fines in sufficient quantity to plug high-efficiency particulate air (HEPA) filters, or (c) cause other conditions, such as excessive dispersion of captured radionuclides or the potential for dust explosions (for combustible dusts). Concentration limits in air for combustible dusts are taken from a report by the US Chemical Safety and Hazard Investigation Board (USCSHIB 2006). Reports and standards from the National Fire Protection Association (NFPA 2015) and the Occupational Safety and Health Administration (OSHA 2005) also are cited as supporting documentation. The specific unit cited in Table 3-2 for mechanical stability (generated fines <420 µm with losses to the off-gas stream of $<50\ \mu g/m^3$) is based upon these known standards for combustible dusts, but it is understood that other issues such as filter plugging and bed compaction may not be measured according to this unit.

Chemical Stability—This metric specifically addresses the effects of chemical species other than the target species. It is assumed that the sorbent is not adversely impacted by the species it is intended to capture.

Regeneration—The desired properties of the sorbents (e.g., high capacity and selectivity) may degrade with each regeneration cycle. Although there is no theoretical limit on how far these properties should be allowed to degrade with each regeneration cycle, limiting degradation to a level that does not markedly worsen plant operation is prudent. A degradation of the desired properties to 80% of their starting values is being set in this evaluation as a minimum acceptable value for this metric. This property is not applicable to sorbents intended for single use.

Decontamination Factor—The DF is a measure of the separation of the target radionuclide from the off-gas stream and other competing components in the off-gas stream under selected operating parameters. It is defined as the flowrate of the isotope in the gas stream entering the capture system, divided by the flowrate of the isotope in the effluent gas stream. When there is negligible change in the inlet and effluent gas flowrates, the DF can be defined as the concentration of the isotope in the inlet gas stream ($[\text{isotope}]_a$) divided by the concentration of the isotope in the effluent gas stream ($[\text{isotope}]_b$). Current US federal regulations for the release of gaseous radionuclides from the nuclear fuel cycle and the resultant estimated radiation doses to the public (EPA 2010, NRC 2012) were used by Jubin et al. (2012a) to determine a set of target DFs for a case study of aqueous nuclear fuel reprocessing facilities. These results are summarized in Table 3-1. Further discussion of the required DF values for a reprocessing facility is outside the scope of this document. The reader is directed to a series of documents that discuss this topic more extensively (Jubin et al. 2011, 2012a, Jubin et al. 2012b, Jubin et al. 2014). Values for the DF depend on the scenario and can range from 1 for ^{3}H and ^{85}Kr (i.e., no abatement required after sufficient cooling of the fuel) to about DF = 8,000 for ^{129}I, which is independent of fuel age. An assessment of target DF values for electrochemical reprocessing facilities is in progress.

Table 3-1. Target DF values for the volatile radionuclides.

Nuclide	DF range
^{3}H	1–700
^{14}C	1–30
^{85}Kr	1–70
^{129}I	1,000–8,000

Co-adsorbed Species—There are “tramp” elements and isotopes that can compete for the same sorption sites as the target radionuclide (e.g., ^{12}C competes with ^{14}C, and chlorine competes with ^{129}I). In some cases, the tramp element forms a thermodynamically more stable compound than the target radionuclide. For some elements, it is impractical to remove or reduce their concentrations without affecting the target radionuclides (e.g., ^{12}C and ^{14}C in CO_2 and ^{127}I and ^{129}I cannot be separated except by isotopic separation techniques). In other cases, co-adsorption can be limited by careful process design. This metric is measured by the mole of each co-adsorbed isotope(s) or element(s) per kilogram of sorbent. For example, if the target species is ^{3}H but the sorbent also co-adsorbs iodine and CO_2, this metric would be the (mol I and mol CO_2)/kg of sorbent. Minimizing the value of this metric is desirable both in terms of the number of species contained within the term and in the total moles of the nontarget species adsorbed.

Flexibility and Pretreatment—The process used to remove the target radionuclide from the off-gas should have a broad operating range to avoid limiting plant throughput through an excessively narrow operating envelope. The operating ranges for such parameters as radionuclide concentration, gas temperature, gas velocity, and other related factors should be as wide as is practicable. Flexibility also refers to the ability of the sorbent to withstand reactive gas phase chemical compounds without significant degradation of its sorption properties.

The concept of gas pretreatment prior to the capture technology is related closely to sorbent flexibility. The less flexible the technology, the greater the likelihood that the gas stream will have to be adjusted before radionuclide removal. There is precedent for the addition of pretreatment steps and other unit operations within capture technologies, but these will increase radionuclide removal costs. Such unit operations also are considered within the process complexity metric, although process complexity takes into account all unit operations, not just those associated with pretreatment.

3.2 Notes on Capture Criteria and Metrics for Target Radionuclides

General notes are provided here on the removal of the four radionuclides expected to require emission control in an aqueous UNF reprocessing facility. Integrated off-gas treatment designs for an aqueous facility are available in Jubin et al. (2016). First, the co-adsorption of ^{129}I should be reduced to the extent possible when capturing ^{3}H to avoid contamination of short-lived ^{3}H radioactive waste with long-lived ^{129}I radioactive waste. Second, if capturing ^{3}H as water (H_2O), the co-adsorption of H_2O from the environment or ambient air must also be considered as part of total system capacity. Similarly, if ^{14}C is to be removed as CO_2, the co-adsorption of $^{12}CO_2$ from the air must be considered as part of total system capacity. Third, it is recommended that the volume of ^{85}Kr-bearing radioactive waste be minimized through the use of noble gas separations that remove xenon from the ^{85}Kr-bearing waste stream. If this separation is not performed, xenon must be considered as part of total system capacity.

3.3 Capture Media Criteria and Metrics

Table 3-2. Capture media criteria and metrics.

Property	Unit	Desired trend	Comments
Metrics for technical performance and physical and chemical characteristics criterion			
Saturation 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 to nontargeted 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 postprocessing 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.
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 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.
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 during both the normal operating and process upset condition temperature ranges is necessary.

Table 3-2. Capture media criteria and metrics (continued).

Property	Unit	Desired trend	Comments
Chemical stability*	% degradation in capacity over time as a function of other species present in gas stream	High stability; low degradation over time	Influences 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 influences 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 nontargeted elements. Tramp elements or isotopes (e.g., Cl, ^{12}C, Xe) fall into this category.
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.

Table 3-2. Capture media criteria and metrics (continued).

Property	Unit	Desired trend	Comments
Process complexity	Number 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., materials safety data sheet, ASTM, Uniform Fire Code, International Fire Code, ANSI, 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, mechanical stability, 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 many customers.
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.

4. WASTE FORMS

Four criteria were identified for waste forms. These are (1) technical performance and characteristics (physical and chemical properties), (2) technical practicality, (3) technical maturity, and (4) cost. For each of these four criteria, multiple metrics were identified that attempt to describe the important aspects of that criterion. Table 4-1 lists the four criteria and the associated metrics. As discussed below, establishing desired values for the waste form metrics is problematic because a geological repository has not been identified. Nevertheless, most metrics are independent of such considerations (i.e., physical, radiological, and chemical durability), or the metrics can be evaluated based using an assumed environment. Other metrics may be required or may need to be revised as progress is made toward the ultimate selection of a disposition pathway.

4.1 Impacts of Waste Form Classification and Repository Requirements

Currently, there is no US candidate repository for high-level nuclear waste and no waste acceptance requirements. Hence, this document attempts to identify the criteria and associated metrics that could be used to select waste forms for the volatile radionuclides. The list of metrics compared favorably with those found in the performance specifications for the proposed Yucca Mountain repository (DOE 2012) (i.e., the Yucca Mountain list was a subset of the list of metrics shown in Table 4-1).

As noted in the introduction, the development of the importance or “value” of each metric within each of the criteria is not included in this document. For the capture metrics and criteria, it is anticipated that the weighting factors for individual metrics may vary with the target isotope. In the case of the criteria and metrics for waste forms, the weighting of the individual metrics and the criteria are expected to be influenced by the specific repository conditions and the waste classification, as well as the specific isotope. For example, the classification of the waste as high-level, low-level, or greater than class C could change the relative importance of a specific criterion. Thus, on a scale of 1 to 10, criterion A might be considered a 4 (moderate importance) if the waste is classified as low-level, but an 8 (relatively high importance) if the waste is classified as high-level. In comparing the performance metrics of two waste forms, it is also critical that these materials be compared with the same repository conditions (i.e., oxidizing or reducing).

4.2 Waste Form Criteria and Metrics

Table 4-1. Waste form criterion and metrics.

Property	Unit	Desired Trend	Comments
Metrics for technical performance and physical and chemical characteristics criterion			
Target element concentration	mol/kg	High	This dictates the amount of waste form that must be made to contain the target radionuclide inventory (moles of isotope per unit mass of the waste form). It influences the total waste form volume, thermal load, curie content, and radiation levels for the containers. This property is based on the elemental concentration of the target species and does not include tramp elements.
Density	kg/m^3	High	The waste form density could be required to determine the size and dimension limits for the waste container or package and is one of the factors that could determine the volume that the waste form occupies during storage, transportation, or disposal.
Heat capacity	$J/(K \cdot kg)$ or $J/(K \cdot m^3)$	Low	Heat capacity controls heating and cooling rates of the waste form when coupled with the heat transfer conditions.
Thermal conductivity	$W/(m \cdot K)$	High	A waste form is heated in two ways—decay heat and co-disposed waste. The waste form must not suffer deleterious property changes because of this heating (see “thermal limits”). Additionally, specifications in the waste acceptance criteria will likely limit the surface temperature and total thermal power of a container.
Mechanical properties	% fines produced	Less than 0.01% fines formation	Dust generation during storage, transport, or disposal, which could be caused by chemical, physical, or thermal changes, should be within expected fines limits.
Chemical properties	Classification according to National Fire Protection Association ratings	Low	Waste forms that display combustibility are strong oxidizers, undergo rapid decomposition, or have the potential for other chemical reactions should be avoided. Included in this metric is the toxicity of the waste form. If the current EPA regulations for hazardous waste apply, the waste form must pass the Toxicity Characteristic Leaching Procedure published by the EPA.
Dissolution or release rate	$g/(m^2 \cdot d)$, $mol/(m^2 \cdot s)$, or equivalent	Low	Upon failure of any applicable engineered barriers, the dose to the public over time from the released radionuclide must be within dose limits. The metric values should be isotope specific (i.e., for iodine), a value such as “less than that for AgI under same redox conditions” might be selected.
Thermal limits	°C	High	Identify an upper temperature limit to prevent excessive property changes in the waste form. A waste form is heated in two ways—internal decay heat and heat from co-disposed waste. The waste form must not suffer deleterious property changes as a result of this heating.
Waste form canister interactions	% alteration of canister or waste form as a function of time	Minimal	The waste form properties cannot be significantly degraded by interaction between the waste form and the engineered barrier materials.

Table 4-1. Waste form criterion and metrics (continued).

Property	Unit	Desired Trend	Comments
Radiation effects	% increase in radionuclide release rate per Gy	High	Identify the maximum dose that the waste form can tolerate. Waste forms must be robust with respect to potential external radiation fields and self-irradiation from the immobilized isotopes. Additionally, as the immobilized isotope decays, a new chemistry evolves. The waste form properties cannot degrade significantly and compromise the repository performance.
Metrics for technical practicality criterion			
Process complexity	Number of control systems and unit operations required	Low	Process complexity plays an important role in the viability of a waste form process. Complex processes might be more difficult to operate and maintain remotely.
Robustness	Maximum % variation in waste form properties without deleterious effects	High	Waste forms that can tolerate wider allowable ranges in properties and still meet waste form performance requirements are preferred. Although process upsets are anticipated, the effect that these have on the overall product should be minimal.
Tolerance to tramp elements	Mol/kg that can be included without deleterious effects	High	During the reprocessing of fuel, tramp elements with similar chemistry to the target radionuclide may be captured and immobilized in the waste form. Understanding their effect on the waste form properties and on the performance of the waste form in a storage facility or repository will be important.
Waste pretreatment	Number of unit operations required to prepare loaded sorbent for waste form production process	Minimal	Ideally, the sorbent can be regenerated, leading to a simple waste stream that can be efficiently processed to a final waste form.
Energy consumption	kW/mol	Low	This is a measure of the amount of energy that is required to covert the captured radionuclide to a final waste form. This considers only the energy consumed within the waste treatment facility and not the energy used in producing the materials consumed.
Metrics for technical maturity criterion			
Technology readiness level	1–9	High	The technology readiness level is defined in the technology readiness assessment report (DOD 2011).
Commercial availability	Yes/No	Available	This can include the commercial availability of a process or of the precursor materials required for waste form manufacture.
Time to commercialization	y	Short	Must be compatible with the time scale of the reprocessing facility.
Metrics for cost criterion			
Cost of materials	$/kg	Low	Cost of consumable materials.
Operating cost	$/Ci	Low	Processing cost less consumables.

4.3 Additional Notes on Waste Form Criteria and Metrics for ^{129}I

One of the most important metrics for waste form performance is dissolution rate. In the case of iodine, because many iodine capture materials capture iodine as AgI, it would be logical to assume that any iodine release rate from a waste form should be lower than that of AgI under expected repository conditions. In testing of potential waste forms, AgI should be included as a reference point because it could represent the direct disposal of the iodine-loaded sorbent.

4.4 Additional Notes on Waste Form Criteria and Metrics for ^{85}Kr

Of the four volatile radionuclides, krypton is unique because it is an inert gas at room temperature. Therefore, there are limited options for converting it to a waste form on which traditional waste form tests can be used. The tabulated metrics for waste forms may need to be interpreted with the understanding that Kr is likely to be stored as a gas in pressurized containers that may or may not contain a filler (e.g., an MOF material or zeolite) that allows more gas to be stored in a container than could be stored in an otherwise empty container at the same pressure. An option is to co-deposit krypton with a metal on the inside of steel containers, thereby locking the krypton in a metal matrix. Several studies have been conducted on the immobilization or encapsulation of xenon or krypton in zeolites (Christensen et al. 1982, Christensen et al. 1983, Kopelevich and Chang 2001, Lim et al. 2001, Miyake et al. 1984, Penzhorn 1981, Penzhorn and Mertin 1984, Penzhorn et al. 1984, Penzhorn et al. 1982, Penzhorn et al. 1980, Seoung et al. 2014, Whitmell et al. 1987). These solid phases could be studied with traditional waste form tests; hence, the metrics for the immobilization solids for the other volatile radionuclides would be important for the krypton immobilization solids.

5. IMPLEMENTATION CASE STUDY

The selective removal of iodine from an off-gas stream can be performed in many ways. To illustrate the use of the criteria and metrics provided in Section 3, iodine removal by $AgNO_3$-coated Berl saddles was compared with iodine removal by silver-exchanged faujasite. These two technologies have both been implemented at engineering scale, which assisted in provision of the information required by the evaluation metric. In Revision 1, the full case study can be found in Appendix B.

Completion of this model case study revealed several important trends. First, not all metrics will apply to any given sorbent system. Second, if sorbent systems are markedly different, the values provided by the metrics may be disparate on first inspection and should be carefully interpreted in the context of the specific technologies being evaluated. Third, the use of general process knowledge or subjective rankings was often required to complete the case study. This is illustrated by the thermal stability metric, where both technologies were designated “good.” Ideally, a more quantitative analysis would be made, but such data were not available. This was a recurring theme in the completion of this case study, and it shows that the performance criteria can be used not only for down-selection between two technologies but also can aid in identifying the knowledge gaps (and their associated importance) that should be resolved during a sorbent or waste form development process.

6. TECHNOLOGICAL DEVELOPMENT OF ^{85}Kr AND ^{129}I REMOVAL PROCESSES

The Materials Recovery and Waste Form Development Campaign within the DOE Office of Nuclear Energy has performed a significant amount of research and development focused on the capture and immobilization of the volatile and semivolatile radionuclides emitted from UNF reprocessing. This section provides an update on specific efforts related to separation of krypton and xenon using solid sorbent technology and on the capture of iodine from plant off-gas streams using silver-based sorbents.

These activities were initiated in 2009 as part of the Off-Gas Sigma Team, a multilaboratory partnership, and continue as of 2020. Contributors have included five national laboratories (Oak Ridge National

Laboratory [ORNL], Idaho National Laboratory [INL], Pacific Northwest National Laboratory [PNNL], Sandia National Laboratory, and Argonne National Laboratory) and universities funded through the Nuclear Energy University Partnership program. A nonexhaustive list of contributing universities includes Northwestern University; University of California, Davis; University of Idaho; Prairie View A&M; Syracuse University; Georgia Institute of Technology; Colorado School of Mines; and Louisiana State University.

Parallel efforts were pursued in the advancement of sorbents for krypton/xenon separations. First, a two-column separative design using an engineered form of mordenite sorbents was developed. Second, the potential for use of MOF materials in the selective removal of krypton and xenon was investigated.

In the case of iodine capture, primary efforts have been focused on two silver-based sorbents. AgZ is a conventional iodine sorbent, and AgAero was considered a novel sorbent at the time research and development was initiated in 2009. Both of these materials have been evaluated for their potential to be directly converted to waste forms for long-term geologic disposal.

This section uses the criteria sets to provide insight into the current status of krypton/xenon separations and iodine removal using these technologies. The intent is to provide an assessment of technological progress, to serve as an information repository, and (most importantly) to identify any remaining barriers to implementation of these technologies at a larger-scale and recommend research to address these outstanding issues.

6.1 Development of Krypton and Xenon Separations

Industrially, the separation of krypton and xenon from air is performed by cryogenic distillation. In air, Kr is significantly more abundant than xenon, with concentrations of 1.14 and 0.087 ppmv, respectively (Haynes 2016). Thus, the engineering design for commercial krypton and xenon recovery will be notably different than the design of a cryogenic distillation process intended for use in nuclear fuel reprocessing, where xenon is produced by nuclear fission at roughly 10× the rate of krypton. Known safety hazards associated with cryogenic distillation include the freezing of xenon in the distillation column, the generation of ozone within the process, and the potential for explosive reactions due to hydrocarbon contamination.

Although cryogenic capture of krypton and xenon in UNF dissolver off-gas streams has been performed, it is a costly and complex process because of the need to operate safely at cryogenic temperatures. The DOG gas stream needs to be purified of contaminants that would otherwise interfere with cryogenic process. Several gas pretreatment unit operations prior to cryogenic distillation as deployed in a UNF reprocessing plant may include selective catalytic reduction (to eliminate NO_X), catalytic oxygen removal, initial H_2O removal through condensation, gas drying (by molecular sieves) and a final cold trap. Following pretreatment, the gas feed may be sent to the distillation columns and associated nitrogen stripping step (Jubin et al. 2016).

From this, it can be seen that cryogenic distillation, as deployed in nuclear fuel reprocessing, is a highly complex process with multiple unit operations, low flexibility, and minimal robustness. It poses several notable safety concerns and is highly energy intensive. When evaluated against the criteria for technical practicality, it scores poorly. These drivers led to the pursuit of solid sorbent alternatives that could potentially be operated at room temperature or at minimum, higher than cryogenic temperatures.

6.1.1 Separation by HZ-PAN and AgZ-PAN

Separation of krypton and xenon using zeolite minerals has been examined by multiple parties (Pence 1981, Monson 1981, Munakata et al. 2003, Jubin et al. 2008). Most recently, INL has developed engineered forms of mordenite minerals using a polyacrylonitrile (PAN) binder. A Ag-exchanged mordenite material (AgZ-PAN) is used for xenon adsorption; a hydrogen mordenite material (HZ-PAN) is used for krypton adsorption (Tables 6-1 and 6-2).

Noble gas separation is effected by passing the xenon- and krypton-bearing feed gas through a column of AgZ-PAN held at 295 K, selectively adsorbing xenon. The gas stream is then passed through a column of HZ-PAN held at 191 K, selectively adsorbing krypton. The noble gases can be removed from the adsorbents by raising the temperature of each column and sweeping each column with inert gas or dry air. The recovered krypton stream is sent to a polishing and purification operation. Although desorption operations have not been optimized, further separation of any krypton co-adsorbed on AgZ-PAN can be achieved through ramped temperature desorption of the AgZ-PAN material.

A conceptual engineering design for separation of xenon and krypton by HZ-PAN and AgZ-PAN was developed by Jubin et al. 2016. In that design, the following unit operations are present:

1. Molecular sieve air dryers
2. AgZ-PAN column for xenon removal (requires heating and cooling)
3. Cooler (to prepare gas stream for 191K HZ-PAN bed)
4. HZ-PAN column for krypton removal (requires heating and cooling)
5. Kr purification
    a. Catalytic recombiner
    b. Cooler
    c. H_2O removal/drying
    d. Cold finger (Kr product condensation)

Advancements in this technology since the conceptual engineering design was developed in 2016 have been focused on thermal cycling of HZ-PAN and AgZ-PAN, defining desorption processes, characterizing breakthrough behavior, and determining the effects that superficial velocity may have on the separation. Relevant references are provided in Tables 6-1 and 6-2.

6.1.2 Separation by Metal Organic Frameworks

PNNL has identified MOFs as promising for the separation of krypton and xenon from the off-gas streams of a used nuclear fuel reprocessing plant. Multiple MOFs have been evaluated, with CaSDB-MOF currently identified as the most promising for room temperature separations (Table 6-1). Some early work also focused on NiDOBDC-MOF (Liu et al. 2014). Efforts continue to identify new materials that may have improved characteristics, and one of the more recently tested MOFs (FMOFCu) has demonstrated selectivity for krypton over xenon at −40°C. If ambient temperature separations are not judged to be efficient, there are low-temperature MOFs (such as CuBTC-MOF, operated at −80°C) that may be useful in krypton/xenon separations.

Broadly, the separation is performed by passing a xenon- and krypton-bearing stream through two column banks both containing the selected MOF. The first column bank removes xenon (with some krypton co-adsorption) and the second removes krypton. Xenon adsorption on CaSDB-MOF occurs at ambient temperatures but is improved at sub-zero temperatures. The noble gases can be removed from the adsorbent by raising the temperature of each column and sweeping each column with inert gas. The recovered krypton is sent to a polishing and purification operation.

Jubin and colleagues developed a conceptual engineering design based on NiDOBDC separation of krypton and xenon. In that design, the following unit operations were present:

1. Molecular sieve air driers
2. Krypton sorbent column bank
3. Xenon sorbent column bank

4. Krypton purification
    a. Catalytic recombiner
    b. Cooler
    c. H_2O removal/drying
    d. Cold finger (krypton product condensation)

Since the conceptual engineering design was developed in 2016, advancements in this technology have been focused on identifying new MOFs with higher noble gas capacities, developing engineered and mechanically robust MOF forms, characterizing breakthrough behavior, defining desorption processes, and identifying other gas components that may co-adsorb with the noble gases. Relevant references are provided in Table 6-1.

6.1.3 Performance Criteria and Metrics for Krypton and Xenon Separation

The following tables reflect the current state of knowledge about krypton and xenon separation through either AgZ-PAN/HZ-PAN or CaSDB-MOF sorbents. Table 6-1 provides metrics for xenon adsorption using either AgZ-PAN and CaSDB-MOF, and Table 6-2 provides metrics for krypton adsorption using HZ-PAN. Not enough data is available to complete the assessment of a MOF material for krypton adsorption. Where possible, technical references are provided. In some cases, the authors' personal experience with the materials and associated unpublished data may be used to qualitatively assess each sorbent.

Table 6-1. Metrics for xenon capture by AgZ-PAN and CaSDB-MOF.

Property	Unit	AgZ-PAN (Xe sorbent)	CaSDB-MOF (Xe sorbent)	Desired trend	Comments
Metrics for technical performance and physical and chemical characteristics criterion					
Capacity (saturated)	mol/m^3	20 mmol/kg [8 mols/m^3] *(295 K)*	19 mmol/kg [12 mols/m^3] *(295 K)*	High	Saturation capacity of both sorbents for noble gases is highly temperature dependent and dependent on sorbate concentration and balance gas composition. The effect of silver content on AgZ-PAN performance is unknown. Breakthrough capacity for each sorbent has been identified as a priority knowledge gap and will be lower than saturation capacity. (Greenhalgh et al. 2016, Welty et al. 2018a, Welty et al. 2018b)
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.*	~35 *(295 K)* Xe/Kr	16 *(295 K)* Xe/Kr	High	The selectivities cited here were obtained with gas blends containing 400 ppm Xe, 40 ppm Kr, and balanced with air. (Garn et al. 2015, Greenhalgh et al. 2015, Welty et al. 2018b, Banerjee et al. 2016)
Surface area	m^2/g	250	81	High	(Garn et al. 2016, Welty et al. 2018a)
Specific heat capacity	J/(K·kg) or J/(K·m^3)	Not yet measured	Not yet measured	Application dependent	
Thermal conductivity	W/(m·K)	Not yet measured	Not yet measured	High	
Radiation stability	% degradation in capacity over time as a function of radiation exposure	Stable	Stable	High stability; low degradation over time	AgZ-PAN has been irradiated to levels as high as 2,500–5,000 kGy; CaSDB-MOF has been irradiated to levels as high as 200 kGy with no observed degradation (unpublished data).

Table 6-1. Metrics for xenon capture by AgZ-PAN and CaSDB-MOF (continued).

Property	Unit	AgZ-PAN (Xe sorbent)	CaSDB-MOF (Xe sorbent)	Desired trend	Comments
Mechanical stability*	Generated fines <420 μm with losses to the off-gas stream of <50 $\mu g/m^3$	None observed	45% <420 um	High stability; low fines generation	(Welty et al. 2018a.) Engineered forms of Ca-SBD have been explored (Thallapally et al. 2019, Fujimoto et al. 2019).
Thermal stability	% degradation in capacity over time at selected operating temperature	None observed	Not yet measured	High stability; low degradation over time	AgZ-PAN experienced 100 operating cycles with no observed sorbent degradation (Welty et al. 2018b). CaSDB-MOF is thermally stable to 400℃ under N_2 flow.
Chemical stability*	% degradation in capacity over time as a function of other species present in gas stream	Stable	Not yet measured	High stability; low degradation over time	AgZ-PAN is reported to withstand degradation by high acid concentrations in radioactive environments.
Reactivity	Compatibility as determined by standardized compatibility tables	Compatible	Not yet assessed	Demonstrated compatibility with all components of gas stream and materials of construction	
Metrics for technical practicality criterion					
Regeneration*	No. of cycles before degrading to 80% of capacity for the target element	>100	Up to 20 cycles	High	The desorption of Xe from both AgZ-PAN and CaSDB-MOF is noted to proceed very slowly. Both AgZ-PAN and CaSDB-MOF experienced 100 operating cycles with no observed degradation (Welty et al. 2018b, Thallapally 2016). The cycle experiments were conducted with inert gases; the effects of a simulated off-gas on regeneration should be evaluated.
Bulk density	kg/m^3	400	633	High	(Jubin et al. 2016, Welty et al. 2018a)

Table 6-1. Metrics for xenon capture by AgZ-PAN and CaSDB-MOF (continued).

Property	Unit	AgZ-PAN (Xe sorbent)	CaSDB-MOF (Xe sorbent)	Desired trend	Comments
Co-adsorbed species*	mmol/kg	Kr, 0.3mmol/kg	Kr, 0.7 mmol/kg 1.2(CO_2) 47(N_2) 5.3(Ar) 12(O_2)	Small in number of species and quantity	Co-adsorption of air components by CaSDB-MOF performed at 295 K with gas streams containing 1,300 ppm Xe and 130 ppm Kr (Thallapally 2017).
Robustness	% variation in operating parameters tolerated without deleterious effects	Not yet assessed	Not yet assessed	High	
Flexibility and pretreatment*	Operating ranges; no. of unit operations for pretreatment	Requires tight temperature control; minimal pretreatment operations.	Requires tight temperature control; minimal pretreatment operations.	High flexibility; minimal pretreatment	In the case of both AgZ-PAN and CaSBD-MOF, variations in temperature will have significant effects on saturation capacity, breakthrough capacity, and other metrics. In the case of AgZ-PAN, superficial velocity was demonstrated to have little effect (Welty et al. 2017a). In Jubin et al. (2016), the only pretreatment step recommended for either AgZ-PAN or MOF-based separations was drying of the noble-gas bearing feed stream.
Process complexity	Number and type of control systems and unit operations required.	Moderate	Moderate	Low	As mentioned in the text, both sorbents are likely to require driers, heating and cooling operations, and Kr purification. Compared to cryogenic distillation, solid sorbent separation designs remain less complex.
Energy consumption	kW/mol	Not yet assessed	Not yet assessed	Low	Both types of sorbents will likely have energy consumption of the same order of magnitude.
Environmental safety and health	Classification according to National Fire Protection Association ratings	Not yet assessed	Not yet assessed	Low	AgZ-PAN will require disposal as a mixed waste. Polyacrylonitrile can form combustible dusts.
Metrics for system design and performance criterion					
Pressure drop	Pa/m	Design dependent	Design dependent	Low	

Table 6-1. Metrics for xenon capture by AgZ-PAN and CaSDB-MOF (continued).

Property	Unit	AgZ-PAN (Xe sorbent)	CaSDB-MOF (Xe sorbent)	Desired trend	Comments
Decontamination factor (DF)*	$[\text{Isotope}]_a/[\text{Isotope}]_b$ (Unitless)	Design dependent	Design dependent	High	Xenon/krypton separation systems should facilitate a plantwide krypton DF of up to 70.
Bed volume	m^3	Design dependent	Design dependent	Low	
Metrics for technical maturity criterion					
Technology readiness level of sorbent system	1–9	6	3	High	Informal assessment.
Commercial availability	Yes/No	No	No	Readily available	AgZ-PAN precursors are readily available.
Time to commercialization	y	1	Not yet assessed	Short	
Metrics for cost criterion					
Cost of sorbent material	\$/kg, \$/Ci, or \$/mol	Not yet assessed	Not yet assessed	Low	
Operating cost	\$/Ci	Not yet assessed	Not yet assessed	Low	

Table 6-2. Metrics for krypton capture by HZ-PAN.

Property	Unit	HZ-PAN (Kr sorbent)	Desired trend	Comments
Metrics for technical performance and physical and chemical characteristics criterion				
Capacity (saturated)	mol/m^3	2.6 mmol/kg [0.9 mols/m^3] *(191 K)*	High	Saturation capacity of HZ-PAN for noble gases is highly temperature dependent and dependent on sorbate concentration and balance gas composition. Breakthrough capacity has been identified as a priority knowledge gap and will be lower than saturation capacity. (Jubin et al. 2016)
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.*	Not yet calculated	High	
Surface area	/m^2/g	330	High	(Greenhalgh et al. 2014)
Specific heat capacity	J/(K·kg) or J/(K·m^3)	Not yet measured	Application dependent	
Thermal conductivity	W/(m·K)	Not yet measured	High	
Radiation stability	% degradation in capacity over time as a function of radiation exposure	Stable	High stability; low degradation over time	HZ-PAN has been irradiated to levels as high as 2,500–5,000 kGy with no observed sorbent degradation (unpublished data).
Mechanical stability*	Generated fines <420 µm with losses to the off-gas stream of <50 µg/m^3	None observed	High stability; low fines generation	

Table 6-2. Metrics for krypton capture by HZ-PAN (continued).

Property	Unit	HZ-PAN (Kr sorbent)	Desired trend	Comments
Thermal stability	% degradation in capacity over time at selected operating temperature	None observed	High stability; low degradation over time	HZ-PAN has been subjected to 50 cycles of operation without observed degradation (Welty et al. 2018b).
Chemical stability*	% degradation in capacity over time as a function of other species present in gas stream	Stable	High stability; low degradation over time	HZ-PAN is reported to hold up to high acid concentration in radioactive environments
Reactivity	Compatibility as determined by standardized compatibility tables	Compatible	Demonstrated compatibility with all components of gas stream and materials of construction	
Metrics for technical practicality criterion				
Regeneration*	No. of cycles before degrading to 80% of capacity for the target element	>50	High	HZ-PAN has been subjected to 50 cycles of operation without observed degradation (Welty et al. 2018b).
Bulk density	kg/m^3	350	High	(Jubin et al. 2016)
Co-adsorbed species*	mol/kg	Not yet measured	Small in number of species and quantity	
Robustness	% variation in operating parameters tolerated without deleterious effects	Not yet assessed	High	

Table 6-2. Metrics for krypton capture by HZ-PAN (continued).

Property	Unit	HZ-PAN (Kr sorbent)	Desired trend	Comments
Flexibility and pretreatment*	Operating ranges; no. of unit operations for pretreatment	Requires tight temperature control; minimal pretreatment operations.	High flexibility; minimal pretreatment	Variations in temperature will have significant effects on saturation capacity, breakthrough capacity, and other metrics. In the case of HZ-PAN, superficial velocity was demonstrated to have little effect (Welty et al. 2017a). In Jubin et al. (2016), the only pretreatment step recommended for either AgZ-PAN/HZ-PAN or MOF-based separations was drying of the noble-gas bearing feed stream.
Process complexity	Number and type of control systems and unit operations required.	Moderate	Low	As mentioned in the text, AgZ-PAN/HZ-PAN separation is likely to require drying, heating and cooling operations, and Kr purification. As compared to cryogenic distillation, solid sorbent separation designs remain less complex.
Energy consumption	kW/mol	Not yet assessed	Low	
Environmental safety and health	Classification according to National Fire Protection Association ratings	Not yet assessed	Low	
Metrics for system design and performance criterion				
Pressure drop	Pa/m	Design dependent	Low	
Decontamination factor (DF)*	$[\text{Isotope}]_a/[\text{Isotope}]_b$ (Unitless)	Design dependent	High	Xe/Kr separation systems should facilitate a plantwide Kr DF of up to 70.
Bed volume	m^3	Design dependent	Low	
Metrics for technical maturity criterion				
Technology readiness level of sorbent system	1–9	6	High	Informal assessment.
Commercial availability	Yes/No	No	Readily available	HZ-PAN precursors are readily available.

Table 6-2. Metrics for krypton capture by HZ-PAN (continued).

Property	Unit	HZ-PAN (Kr sorbent)	Desired trend	Comments
Time to commercialization	y	1	Short	Informal assessment. The actual time depends on continuing research and development funding levels and implementation in a pilot or demonstration-scale facility that could increase the TRL.
Metrics for cost criterion				
Cost of sorbent material	\$/kg, \$/Ci, or \$/mol	Not yet assessed	Low	
Operating cost	\$/Ci	Not yet assessed	Low	

6.1.4 Recommended research pathways for krypton/xenon separations

Upon review of available data and consultation with subject matter experts, a list of outstanding research questions were identified on the topic of krypton/xenon separations using engineered zeolites or MOF adsorbents. Although there are many fundamental studies that could be performed, the work proposed here is judged as providing high value to overall technology advancement. The engineered zeolites, as evidenced by the documented metrics, are at a higher level of development than the MOFs; consequently, questions of a more elementary nature were identified as important to the advancement of MOF materials for krypton and xenon capture and separations.

6.1.4.1 Kr/Xe separations using engineered zeolites

First, the desorption processes for AgZ-PAN and HZ-PAN have not been optimized. To date, the majority of desorption experiments have been performed with a helium sweep gas (Welty et al. 2017b). Preliminary studies indicate that desorption using air is feasible for the removal of xenon from AgZ-PAN but further experimentation to confirm and refine xenon desorption methodology is recommended. Importantly, desorption processes for krypton also require additional design and refinement. Design and testing of krypton desorption processes will support an understanding of the product purity that can be obtained via engineered zeolite separations and whether additional polishing steps (such as condensation or additional adsorption stages) will be required after the initial krypton/xenon separation.

Second, rate data for adsorption of krypton and xenon by the engineered zeolites has not yet been collected. This data was identified by Jubin et al. (2017b) as augmenting the metrics included in Table 3-2 under the technical performance and physical and chemical characteristics criterion. In the case of capture technologies functioning by physisorption, the adsorption rate will dictate the bed depth required to obtain target removal efficiencies. The effect of co-adsorbed species (including N_2, O_2, and other air components) on the adsorption rate and material capacity should be assessed.

Third, completion of irradiation studies is recommended. These studies were previously initiated but concluded prematurely when resources were refocused to other experimental needs. Zeolite minerals are largely resistant to radiation-induced degradation and validation that the engineered zeolite forms retain this characteristic is suggested.

Finally, a technical review of krypton/xenon separation by AgZ-PAN/HZ-PAN is recommended. This review should synergize the work performed to date, including the refinement of the engineering design developed by Jubin et al. (2016), to assess the cost savings and operational benefits that may be realized from implementation of this technology in place of cryogenic distillation. The technical data collected by the Off-Gas Sigma Team since 2009 should facilitate the execution of this review at a reasonable level of fidelity.

6.1.4.2 Kr/Xe separations using MOFs

To date, the separation of krypton and xenon using MOFs has been focused on deploying a two-column adsorbent system, similar to the deployment scenario envisioned for engineered zeolites in which the first column is optimized for xenon removal. This is certainly a potential scenario, but the ideal separation would be performed using a single column system with a MOF selective for krypton over xenon. FMOFCu has been identified as selective for krypton over xenon at -40°C, but limited data is available on this material. A robust research effort would work to identify additional krypton-selective MOFs designed to operate at ambient or near-ambient temperatures. This search will benefit from the tunable nature of MOFs and the large research community interested in these materials for diverse applications. Both experimental and computational studies to support identification of promising materials should be supported.

In the case of CaSDB-MOF, several key issues require resolution. First, breakthrough capacity for CaSDB-MOF in prototypical conditions should be determined. Preliminary research indicates that breakthrough may be near immediate, which would present substantial obstacles in the design of a separative system. This issue is potentially resolved through selection of a lower operating temperature, but an optimization between the operating temperature and xenon capacity has not yet been performed. Second, development of desorption methodology should be performed to assess regeneration time, energy requirements, and potential product concentration and purity. Finally, irradiation studies are of high priority for MOF materials, as their organic structural components may make them susceptible to radiolytic degradation. Results from preliminary irradiation studies are promising, as shown in the metrics for CaSDB-MOF.

6.2 Iodine Sorbent Development

6.2.1 AgZ and AgAero

AgZ has been studied in detail as an iodine adsorbent. Like silver faujasite (described in Section 5), iodine is chemisorbed by silver present in the material to form AgI. Recent research by the Off-Gas Sigma Team at both ORNL and INL, along with university colleagues, has focused on the resistance of AgZ to the various components present in the off-gas streams generated by nuclear fuel reprocessing (such as reported in Greaney and Bruffey 2020), its ability to adsorb organic iodides (Jubin et al. 2017), and characterization of fundamental engineering data (such as reported in Haefner and Soelberg 2009, Soelberg et al. 2013, Soelberg 2013, and Soelberg and Watson, 2016). These studies have significantly advanced the ability to design an iodine removal system using AgZ. Concurrently, development of an alternative silver-based sorbent, silver-functionalized silica aerogel (AgAero) was initiated by PNNL and advanced through collaborations with ORNL and INL. Similar development efforts to those of AgZ (including assessing the resistance of AgAero to degradation by gas stream components, collection of engineering data, and characterizing the adsorption of organic iodides) were pursued (such as reported in Matyáš et al. 2018, Soelberg and Watson 2016, Jubin et al. 2017a, Soelberg et al. 2019). Additionally, PNNL has focused on improving the mechanical stability of AgAero, which was identified early as a barrier to use.

The deployment scenario for both AgZ and AgAero is identical. The expected operating temperature is 150°C, and both sorbents are expected to require limited pretreatment of the iodine-bearing feed stream. Both sorbents are considered single-use and are destined for disposition following use, prompting studies to determine their suitability for direct conversion to an iodine-bearing waste form, discussed further in Section 6.3 (Jubin et al. 2017b, Matyáš et al. 2016).

6.2.2 Performance Criteria and Metrics for Iodine Sorbents

Table 6-3 reflects the current state of knowledge of iodine capture using either AgZ or AgAero. Iodine capture is assumed to be conducted under prototypic DOG conditions, in which the gas stream is likely to contain primarily elemental iodine (I_2) in a humidified gas stream. The gas stream is also expected to contain 1–2 vol% NO and NO_2, and iodine concentrations are expected to be in the parts-per-million concentration range. Where possible, technical references are provided. In some cases, the authors' personal experience with the materials and associated unpublished data may be used to qualitatively assess each sorbent.

Table 6-3. Metrics for iodine capture by AgZ and AgAero.

Property	Unit	AgZ (Iodine sorbent)	AgAero (Iodine sorbent)	Desired trend	Comments
Metrics for technical performance and physical and chemical characteristics criterion					
Capacity (saturated)	mol I/m^3	≈ 800	≈ 1400	High	Maximum capacity based on sorbent volume is dependent on silver content and bulk density of the adsorbent, which can vary. Typical silver content for AgZ is 100 mg/g. The majority of recent testing on iodine uptake was performed using AgAero having a silver content of ≈ 250 mg/g, roughly corresponding to an iodine capacity of 1,400 mol I/m^3.
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.*	See comments	See comments	High	Silver-based sorbents may react with and chemisorb halogens and oxygen. Other halogens and oxygen compete with iodine chemisorption. Although not specifically evaluated in tests, other halogens are believed to compete with iodine in proportion with their relative gas stream concentrations, reducing the sorbent capacity for iodine in proportion with their relative gas stream concentrations. For additional information on sorbent oxidation, see Chemical stability.
Surface area	m^2/g	Not yet measured	120	High	
Specific heat capacity	J/(K·kg) or J/(K·m^3)	Not yet measured	Not yet measured	Application dependent	
Thermal conductivity	W/(m·K)	Not yet measured	Not yet measured	High	
Radiation stability	% degradation in capacity over time as a function of radiation exposure	Not yet measured	Not yet measured	High stability; low degradation over time	Zeolites are relatively stable when exposed to radiation (e.g., Lopez et al. 1995). The silica aerogel support used in manufacture of AgAero is stable when exposed to radiation (Sahu et al. 1996).

Table 6-3. Metrics for iodine capture by AgZ and AgAero (continued).

Property	Unit	AgZ (Iodine sorbent)	AgAero (Iodine sorbent)	Desired trend	Comments
Mechanical stability*	Generated fines <420 µm with losses to the off-gas stream of <50 µg/m^3	High	Low	High stability; low fines generation	The specific unit cited here is based upon standards for combustible dusts (USCSHIB 2006, NFPA 2015, and OSHA 2005). Although AgZ and AgAero fines are not expected to be combustible, these values are used for this metric for AgZ and AgAero until more applicable values based on sorbent mass loss, captured radionuclide loss, or impact on downstream equipment are determined. Commercially available AgZ has high mechanical stability (Puppe and Wilhelm 1990). AgAero produces >50% fines <354 µm in the presence of 1 vol% NO_2 (unpublished). This level of fines generation is likely to increase iodine mobility from the sorbent bed or contribute to column plugging. Efforts are ongoing to improve the mechanical stability of this material (Matyáš et al. 2019).
Thermal stability	% degradation in capacity over time at selected operating temperature	High	High	High stability; low degradation over time	No observed degradation in capacity attributable to thermal degradation for either sorbent during 4 months at operating temperature of 150°C (Jubin et al. 2017).
Chemical stability*	% degradation in capacity over time as a function of other species present in gas stream	Moderate	Limited	High stability; low degradation over time	AgZ capacity is reduced by 60% when exposed to a moist air stream for 4–8 months (Greaney and Bruffey 2020) and by 30% percent when exposed to a stream containing NO_2 for 1 week (unpublished). AgAero degrades upon exposure to NO_2 (see Mechanical stability) and experiences capacity loss in humid (22% loss in 6 months) and NO_2 bearing gas streams (15% capacity loss in 4 months) (Matyáš et al. 2018). Loss of capacity for both sorbents is presumed to be due to oxidation of metallic silver to silver oxide species. (See Selectivity.) A substantial amount of published data on this topic exists, and the reader is referred to that for additional information (Huve et al. 2018).

Table 6-3. Metrics for iodine capture by AgZ and AgAero (continued).

Property	Unit	AgZ (Iodine sorbent)	AgAero (Iodine sorbent)	Desired trend	Comments
Reactivity	Compatibility as determined by standardized compatibility tables	See comments	See comments	Demonstrated compatibility with all components of gas stream and materials of construction	AgZ and AgAero are compatible with typical materials of construction and relevant gas streams.
Metrics for technical practicality criterion					
Regeneration*	No. of cycles before degrading to 80% of capacity for the target element	n/a	n/a	High	Silver-sorbents can theoretically be regenerated but are typically considered single-use.
Bulk density	kg/m^3	≈ 850	≈ 600	High	Independent measurements.
Co-adsorbed species*	mol/kg	Halogens; H_2O	Halogens	Small in number of species and quantity	Silver-based sorbents will capture other halogens present in the gas streams to form AgCl, AgBr, etc. AgZ will adsorb 0.5 wt% H_2O (including tritiated H_2O) at 150°C (Spencer et al. 2013).
Robustness	% variation in operating parameters tolerated without deleterious effects	Not yet examined	Not yet examined	High	
Flexibility and pretreatment*	Operating ranges; no. of unit operations for pretreatment	Limited pretreatment required; flexibility not yet examined	Limited pretreatment required; flexibility not yet examined	High flexibility; minimal pretreatment	The gas stream must be heated before entering either the AgZ or AgAero sorbent bed because both sorbents are expected to operate at >100°C (Jubin et al. 2016).
Process complexity	Number and type of control systems and unit operations required.	Low	Low	Low	Unit operations and control systems are likely to be comprised of (1) gas stream preheating, (2) adsorbent beds, (3) iodine monitor, (4) gas stream cooling (if needed) (Jubin et al. 2016). No differences in process complexity are expected between AgZ and AgAero.

Table 6-3. Metrics for iodine capture by AgZ and AgAero (continued).

Property	Unit	AgZ (Iodine sorbent)	AgAero (Iodine sorbent)	Desired trend	Comments
Energy consumption	kW/mol	Not yet assessed	Not yet assessed	Low	AgZ and AgAero are expected to have similar operational energy consumption. The pressure drop across sorbent bed(s) imposes energy demand by induced draft fan(s), but such pressure drop is expected to be in a normal range for off-gas control equipment of less than 10–20 in. H_2O.
Environmental safety and health	Classification according to National Fire Protection Association ratings	Low	Low	Low	Silver-based sorbents will be classified as Resource Conservation and Recovery Act waste unless they have been exposed to radioactive off-gas streams, in which case they would be classified as mixed waste.
Metrics for system design and performance criterion					
Pressure drop	Pa/m	Design dependent	Design dependent	Low	See Energy consumption.
Decontamination factor (DF)*	$[\text{Isotope}]_a/[\text{Isotope}]_b$ (Unitless)	Design dependent	Design dependent	High	Iodine adsorbents should be capable of achieving DFs of 1,000–8,000 (Jubin et al. 2012a). Testing to date has shown that this range of DFs is generally achievable even in cases where the sorbent capacity is depressed because of aging.
Bed volume	m^3	Design dependent	Design dependent	Low	
Metrics for technical maturity criterion					
Technology readiness level of sorbent system	1–9	5	3	High	Informal assessment.
Commercial availability	Yes/No	Yes	No	Readily available	AgAero is currently manufactured at gram scale. Precursors and required reagents are readily available.
Time to commercialization	y	<3	<5	Short	Informal assessment of earliest possible implementation. The actual time depends on research and development funding levels and implementation in a pilot or demonstration-scale facility that could extend the TRL to 6 or higher.

Table 6-3. Metrics for iodine capture by AgZ and AgAero (continued).

Property	Unit	AgZ (Iodine sorbent)	AgAero (Iodine sorbent)	Desired trend	Comments
Metrics for cost criterion					
Cost of sorbent material	\$/kg, \$/Ci, or \$/mol	Moderate	Moderate	Low	The driving cost for silver-based sorbents is the silver-bearing chemicals used in production.
Operating cost	\$/Ci	Low	Low	Low	The operating cost for both sorbents is expected to be low because of the minimal number of required unit operations, simple design, low energy costs, and the lack of significant health and safety hazards.

6.2.3 Recommended research pathways for iodine sorbents

As Table 6-3 shows, the development status of AgZ and AgAero are similar, with two notable exceptions. First, the mechanical stability of AgAero has not yet been confirmed as sufficient for use in the target off-gas streams. AgAero is observed to degrade upon extended exposure to moisture-bearing gas streams (Jubin et al. 2017a) and degrades rapidly upon exposure to 1 vol% NO_2-bearing gas streams (unpublished). For this reason, PNNL is developing a physical form of AgAero that will be more resistant to degradation from expected gas stream components. Second, AgAero is not yet commercially available or produced at large scale. There are no immediately presenting barriers to commercial production of AgAero.

Outstanding research questions pertaining to both iodine sorbents have been well-developed in several documents authored by the Off-Gas Sigma Team in recent years. These documents include an engineering evaluation of an integrated off-gas treatment system (Jubin et al. 2016), identification of data requirements for pilot-scale deployment of an integrated off-gas treatment system (Jubin et al. 2017b), and development of a test plan focused on fundamental questions surrounding organic iodine adsorption by silver-based sorbents at very low iodine concentrations (Jubin et al. 2018).

Many of the knowledge gaps identified for iodine capture by AgZ and AgAero are related to the adsorption of iodine from streams other than the DOG. Of high priority are the VOG and vitrification off-gas streams. The VOG is expected to contain a high proportion of organic iodides at very low concentrations (likely parts per billion). As detailed in Bruffey et al. (2015), very little is known about the adsorption of organic iodides by silver-based sorbents at these concentrations. Initial efforts have been conducted at both ORNL and INL, but efforts are limited because of the level of resources required to address the overarching questions associated with this topic. Specifically, the time required to conduct these types of adsorption tests and the analytical instrumentation and testing needed make it challenging to sustain an experimental program. However, the issuance of the test plan referenced previously (Jubin et al. 2018) has been beneficial in coordinating experimental priorities, and meaningful progress can still be achieved. Seven key questions were identified in that document, and they are included here for convenience. These questions demonstrate that needed research on this topic is highly fundamental in nature. Some work is ongoing at ORNL and INL to address these questions, and future work should expand this experimental program (Bruffey et al. 2019).

1. How are the adsorption rates of organic iodides by silver-based sorbent related to hydrocarbon chain length?

2. Are the adsorption rates a function of long-chain organic iodide concentration?

3. What is the effect of the gas velocity on the behavior observed in questions 1–2?

4. What is the saturation concentration of iodine for various long-chain organic iodides on silver-based sorbents, and does it vary with target feed species?

5. What is the DF over a fixed length of bed as a function of concentration and iodine species in the feed gas?

6. If the adsorption rate changes, which is impacted the most: the DF, bed penetration depth, or bed penetration rate? (This is the combined effect of the results of questions 1, 2, 3, and 5.)

7. What is the length and shape of the MTZ, and how do they vary or change for CH_3I and other organic iodides on silver-based sorbents?

In addition to efforts on iodine capture from VOG streams, limited research is available on iodine capture from waste processing operations, including iodine capture from the vitrification off-gas. Likely, the vitrifier will emit primarily inorganic iodine species (I_2, ICl, IF_x, etc.), and other gas stream properties (such as composition, flowrate, and temperature) will be markedly different from DOG and VOG. An analysis of this gas stream should be conducted to better understand this potential application of iodine sorbents.

6.3 Progress on Iodine Waste Form Development

A review of available iodine waste forms is available in Riley et al. (2016). Since publication of that review, substantial advancements have been made in the field. Many of these advancements have been supported by DOE Office of Nuclear Energy through direct funding or through university grants within the Nuclear Energy University Partnership. Revision 1 of this document summarizes these updates. For most of the waste forms discussed here, an evaluation using the developed criteria and metrics is not meaningful, given their early stage of development. Diverse iodine waste form research and development should continue and promising forms should continue to be advanced to higher levels of development. Included here are iodine waste form options for both iodine immobilization from gaseous and aqueous waste streams.

A summary of some of the recent waste form options for iodine is provided in Table 6-4. The waste forms listed are updates from the recent literature, most of which are in addition to the previous summaries provided in previous reviews (Jubin et al. 2016, Riley et al. 2016). These will be described in more detail in subsequent sections.

Table 6-4. Summary of iodine capture options linked to waste forms (Asmussen et al. 2019b, Lawter et al. 2019).

Waste form	Specifics	Iodine form	Iodine loading (mass%)	Reference
Glass-bonded ceramics	AgZ+I (HIP)	AgI	3–13.5%	Bruffey et al. (2017), Jubin et al. (2017c), Bruffey and Jubin (2018), Asmussen et al. (2019b),
	Iodosodalite (NaI)	NaI	22%	Chong et al. (2018), Nam et al. (2018), Riley et al. (2019)
	Iodosodalite (HIP) (NaI)	NaI	18–20%	Chong et al. (2020)
	Iodosodalite (AgI)	AgI	14%	Riley et al. (2019)
	Iodoapatite	PbI_2	7%	Yao et al. (2014), Zhang et al. (2018)
Glass waste forms	Tellurite glass	AgI	12.6%	Lee et al. (2017)
Aerogels	AgAero+I (HIP) AgAero+I (SPS)	AgI	30%	Matyáš and Engler (2013), Matyáš et al. (2018), Asmussen et al. (2019b)

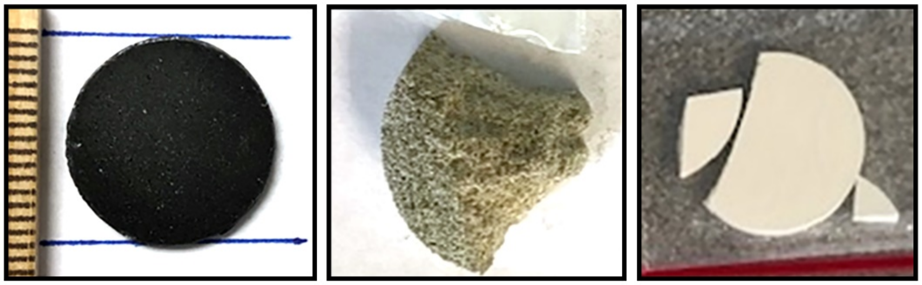
Note: SPS denotes spark plasma sintering, and HIP denotes hot isostatic pressing.

A summary of the waste form appearances for those listed in Table 6-4 are provided in Figure 6-1 with scanning electron micrographs of the waste forms before corrosion testing shown in Figure 6-2. Some of the waste forms, including the enclosure, can used for hot isostatic pressing (HIP).

Large-format Ag^{0}Z (low I) (ORNL)

Large-format Ag^{0}Z (high I) (ORNL)

Ag^{0}Z (ORNL)



Ag^0-silica aerogel (HIP and SPS) (PNNL)

Glass-bonded sodalite ceramics (PNNL/INL)

Iodosodalite (WSU-PNNL NEUP)

Iodosodalite (U. of Birmingham)

Tellurite glass (POSTECH)

Pb-V iodoapatite (LSU)

Figure 6-1. Samples evaluated during the PNNL testing of iodine waste forms. This figure was modified from the original published by Asmussen et al. (2019b) and Lawter et al. (2019).

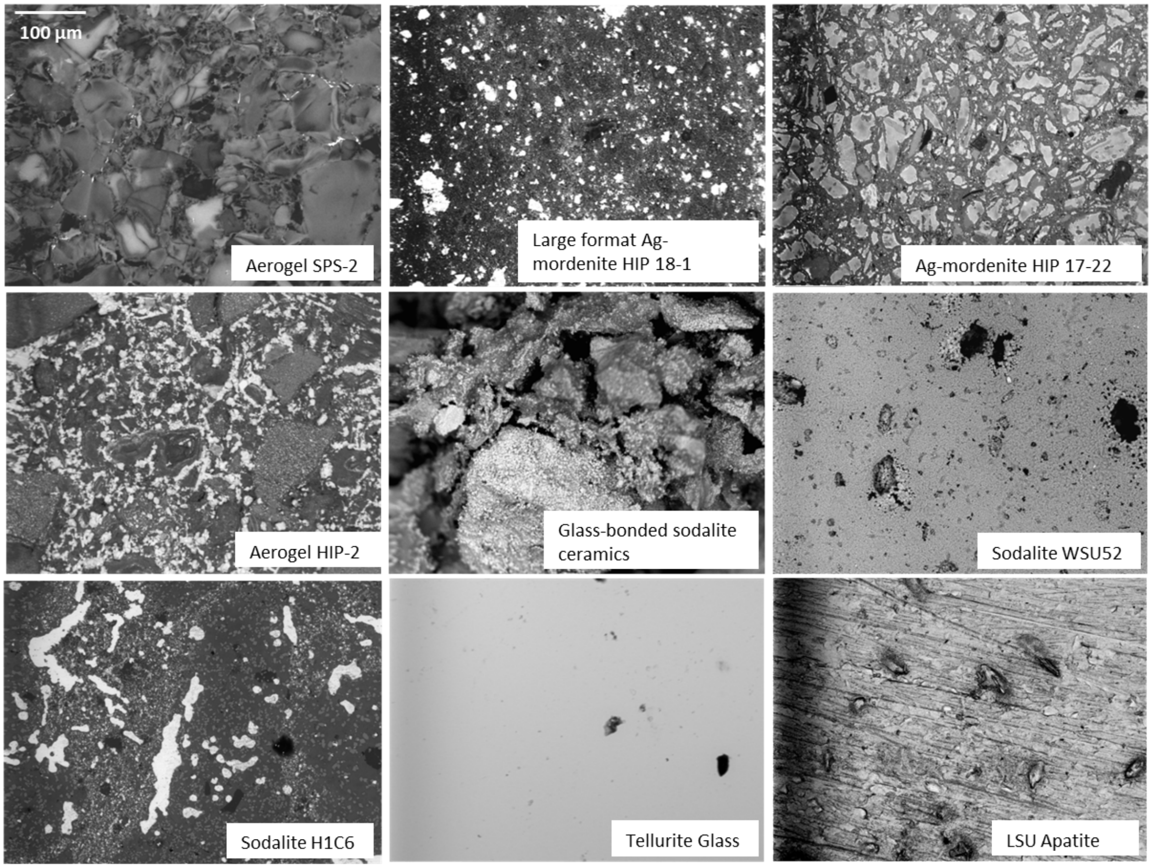


Figure 6-2. SEM micrographs of the various sample types used in this study before corrosion testing. All images were collected at 250×; the scale bar in the top left corner applies to all SEM micrographs. These were taken from Lawter et al. (2019).

6.3.1 Hot Isostatically Pressed Iodine-Loaded Ag-Mordenite (Ag^{0}Z+I) Waste Forms

Direct conversion of iodine-bearing zeolites into a waste form using HIP has been investigated by several researchers, and ORNL has recently extended these efforts. HIP processing of iodine-loaded silver zeolite or silver silica gel with a metal is described in a Japanese and US patent (Fukumoto 1998), which mixed either iodine-loaded faujasite or iodine-loaded silver-impregnated silica gel with 50 vol% copper powder and pressed the samples at 860°C at 195 MPa for 3 h. This resulted in confinement of AgI particles to the adsorbent, and the encapsulation of the adsorbent within a sintered metallic matrix. A second Japanese study HIPed silver nitrate–impregnated alumina at 1,200°C and 175 MPa for 3 h. The resulting product had a density greater than 4.0×10^3 kg/m^3 (Tanabe 2010). There are two reported conversions of zeolites to sodalite using HIP. In 2005, the National Nuclear Laboratory in the United Kingdom demonstrated HIP conversion of A and X zeolites at 900°C, converting them to a sodalite (Maddrell 2005). Sheppard et al. (2006) investigated the conversion of various iodine-bearing silver zeolites to form I-sodalite. These included the silver-exchanged zeolites of structural type A, X, and Y. These tests were conducted with powdered zeolites occluded with powdered AgI. HIP conditions were 900°C at 190 MPa for 2 h. Silver zeolite Y did not form sodalite, whereas silver-exchanged zeolites A and X appear to form monolithic sodalites.

Efforts at ORNL have specifically focused on the conversion of AgZ to a waste form using HIP. Initial scoping studies were conducted by hot uniaxial pressing and were followed by more systematic HIP studies identifying pressing conditions, the mineral phases produced during HIPing, and the homogeneity of iodine distribution within the pressed mineral (Bruffey and Jubin 2016, Jubin and Bruffey 2015). The most recent experimental study conducted by ORNL on the HIPing of I-AgZ focused on multiple mordenite forms, including sodium mordenite (NaZ), pure AgZ, and engineered AgZ that includes a clay binder material (Bruffey and Jubin 2015). The variables investigated included the pressure and temperature of pressing, the ratio of mineral to iodine, and the form of iodine (NaI vs. AgI). The minerals and iodide forms were ground together by mortar and pestle and then pressed using the desired pressing conditions. The pressed samples were characterized according to their densities and analyzed by x-ray diffraction to ascertain mineral phases, x-ray diffraction revealed that after pressing, the sample material was largely amorphous, with some SiO_2 and AgI phases distinct within the sample. These results indicate that HIP of I-AgZ is likely to produce a durable, densified waste form (Bruffey et al. 2016).

6.3.2 Glass-Bonded Iodosodalite

Iodosodalite has been studied for several decades for the immobilization of iodine (Strachan and Babad 1979, Barnes et al. 1983, Maddrell et al. 2015, Chong et al. 2018, Nam et al. 2018, Riley et al. 2019, Chong et al. 2020). Recent work has involved some different approaches to generating the precursors for iodosodalite waste form production. The general formula for NaI-sodalite is $Na_8(AlSiO_4)_6I_2$, but sodium can be substituted for other 1+ cations. In a study by Chong et al. (2018), iodosodalite was made via a hydrothermal process at 70°C starting with NaI as the iodine source. In a related study, Nam et al. (2018) made iodosodalite using similar precursors, but this work was done in an aqueous (nonhydrothermal) process at 70°C using NaI as the iodine source. Taking the most promising products from the Chong et al. (2018) and Nam et al. (2018) studies, glass-bonded iodosodalite waste forms were fabricated using HIP (Chong et al. 2020). These waste forms showed moderate iodine release rates with the best results coming from the aqueously produced precursors mixed with 20 mass% of a glass binder.

In a separate study, glass-bonded iodosodalite was made using a dry process with zeolite 4A and either NaI or AgI (Riley et al. 2019). In both cases, samples were made in an argon glovebox with low moisture content to prevent zeolite hydration. Salts were occluded in the zeolite 4A at 665°C (NaI) or 600°C (AgI) for 24 h. Then, 26 mass% of a glass binder was added to each, the mixtures were loaded into glassy carbon crucibles, and heat-treated at 925°C for 24 h. The final products had high porosities. The final NaI-sodalite product was pure iodosodalite, whereas AgI-sodalite product did contain some sodalite but also a lot of nepheline, AgI, and even some Ag^0. If a process such as this were to be used in the future for making iodine waste forms, several options are available for product improvements including additional (or more optimized) glass binders and pressure-assisted sintering to help densify the waste forms as well as help prevent iodine loss during the sintering process.

6.3.3 AgI-Tellurite Glass

In a study by Lee et al. (2017) a Te–Ag–Bi–O tellurite glass was made to immobilize AgI. This glass was of the composition $41.3TeO_2–25.7Ag_2O–11.3Bi_2O_3–21.7AgI$ (by mass). To make this glass, powders of these additives were mixed together and melted in alumina crucibles at 700°C for 30 min. It was determined that 96% of the iodine remained after the consolidation process with 12.64 mass% of iodine loaded in the final waste form. The iodine normalized release for this waste from was very low at 6.5×10^{-4} g/m^2 using the product consistency test.

6.3.4 Ag^0-Functionalized Silica Aerogel (AgAero) Waste Forms

The Ag^0-functionalized silica aerogel (AgAero) has been demonstrated to capture iodine from gaseous streams as well as aqueous streams (i.e., as I^- or IO_3^-) (Matyáš et al. 2011, Matyáš and Engler 2013, Matyáš et al. 2018, Asmussen et al. 2019a). These materials are made by installing thiol (i.e., –SH) tethers to silica aerogels, adding Ag^+ to the thiol layers using a soak in $AgNO_3$ solution, followed by reduction of the Ag^+ to Ag^0 under a flow of Ar/H_2. Recent work has been done to evaluate the properties of consolidated iodine-loaded AgAero sorbents using HIP, spark plasma sintering, and hot uniaxial pressing. All of these consolidated waste forms were taken to near-theoretical densities with low residual porosities. Removing the organic portion of the sorbent from the installation of the thiol tethers, as well as adding additional silica (i.e., colloidal silica) helped improve the densities of the final consolidated products.

6.3.5 Chemical Durabilities

To assess the durability of iodine waste forms for long-term disposal, a predictive model is required. In order to populate said model a test suite is required that can provide relevant durability data. An ideal test suite would include a rapid screening of iodine waste forms durability to assess improvements during material development coupled with a time- and cost-effective longer-term test to assess the overall dissolution mechanisms. Several test methods have been considered previously including ASTM C1662-10 single-pass flow-through test (Asmussen et al. 2019), ASTM C1285-14 product consistency test (Jubin and Bruffey 2015), and modified ANSI/ANS 16.1 testing (Lawter et al. 2019).

Recent work at PNNL and ANL funded through DOE Office of Nuclear Energy has focused on defining a suitable test suite for iodine waste form evaluations and determining the controlling dissolution mechanisms of various iodine waste form types (Ebert 2019, Ebert et al. 2019, Asmussen et al. 2019, Lawter et al. 2019.) The test suite being developed is comprised of two portions: (1) a rapid, single interval leach test (~3 d long) that can be used to screen iodine waste form samples of the same material type in development efforts and (2) a dynamic leach test (~14 d or longer) that can be used to acquire dissolution rate data to be used to develop and populate a predictive durability model. A key feature of the test suite is the use of monolithic iodine waste form samples that allow for observations of the evolution of the iodine waste form microstructure during and after the test. This data can be used to understand the mechanisms controlling iodine release from the iodine waste forms.

An example data set showing the durability response of several iodine waste form samples subjected to the 3 day screening leach test is shown in Table 6-5. The screening test was designed to compare samples of the same material class and shows that the small form AgZ samples were more durable than the large form, spark plasma sintering AgAero was more durable than HIP AgAero and the iodosodalites had wide variable durability within sample sets. Further information and analysis can be found in Lawter et al. (2019).

Table 6-5. Normalized dissolution rate (NL_I), with respect to iodine, for the various samples evaluated in the 3 day leach test in Lawter et al. (2019). Note that this is a summary of that data and includes a range of values for each type of waste form evaluated. The term <EQL means that this was below the instrument detection limits.

Iodine Waste Form	NL_I (g/m^2/d)
Ag-mordenite (large form)	0.132–0.505
Ag-mordenite (small form) (ORNL)	0.014–0.08
Ag^0SA (HIP) (PNNL)	0.387
Ag^0SA (SPS) (PNNL)	0.001
Iodosodalite (UK)	0.002–40.1
Iodosodalite (WSU)	1.11–192
Iodosodalite (PNNL-NaI)	32.8
Iodosodalite (PNNL-AgI)	674
Iodoapatite ceramic (LSU)	3.25
Tellurite glass	<EQL

Note: SPS = spark plasma sintering.

7. SUMMARY AND CONCLUSIONS

While compiling these criteria and associated metric sets, many observations were made. First, the use of the tables in this document should include the following considerations:

1. The tables, as presented, do not weight the importance of one criterion against another. Different users could have different perspectives about the relative importance of each criterion.
2. The tables also do not weigh the relative importance of individual metrics associated with a particular criterion.
3. Some specific metrics may not be applicable for comparison if the capture or waste form technologies being compared differ substantially (e.g., comparing the density of a wet scrubber system with that of a solid sorbent is not very informative).

Second, it is important to recognize that some of the metrics listed may be interrelated in complex ways. For example, waste loading and waste density both impact total waste volume, and high density could compensate for low waste loading (mol/kg). This type of relationship should be acknowledged when comparing sorbent systems or waste form technologies.

One of the more complex aspects of this effort to identify the key criteria and metrics is that, as in many engineering problems, there is not a single answer, and some of the important metrics are actually a function of the system design and not an intrinsic property of the material When using these metrics, it is important to use values from properly designed systems.

Third, while completing the case study for silver-based iodine sorbents, quantitative data were not available for many of the performance metrics identified in Section 3 (or Section 4), which leads to the important point that some data gaps remain that should be addressed in the sorbent or waste form development process. At all points of the research and development process, the importance of a metric can be used to identify research priorities for each sorbent or waste form. Additionally, down-selection of sorbents or waste forms is likely to occur at multiple levels of development, and these performance metrics are expected to contribute positively to that determination.

In Revision 1, the status of krypton/xenon separations using engineered zeolite minerals and MOF materials was assessed using the criteria available in Table 3-2. It was found that the zeolite-based separation is relatively advanced in its development, but several key issues require resolution. First, desorption processes for both krypton and xenon require additional design and refinement. This will provide an understanding of the product purity that can be obtained via engineered zeolite separations and whether additional polishing steps (such as condensation or additional adsorption stages) will be required following the initial krypton/xenon separation. Second, adsorption rate data for adsorption of krypton and xenon by the engineered zeolites has not yet been collected. This data is critical in the calculation of required bed depth to achieve a specified product purity. Finally, it is strongly recommended that a technical review of krypton/xenon separation by AgZ-PAN/HZ-PAN be performed. This review should synergize the work performed to date, including the refinement of the engineering design developed by Jubin et al. (2016) to assess the cost savings and operational benefits that may be realized from implementation of this technology in place of cryogenic distillation.

Assessment of MOFs for their use in the separation of krypton/xenon found that the ideal separation would be performed using a single column system with a MOF selective for krypton over xenon. A robust research effort should work to identify multiple krypton-selective MOFs designed to operate at less-than-cryogenic or ambient temperatures. This search will benefit from the tunable nature of MOFs and the large research community interested in these materials for diverse applications. Both experimental and computational studies to support identification of promising materials should be supported. For CaSDB-MOF (the most well-understood xenon sorbent to date), two issues are judged of high importance. First, xenon breakthrough capacity for CaSDB-MOF in prototypical conditions should be determined.

Preliminary research indicates that breakthrough may be near immediate, which would present substantial obstacles in the design of a separative system. Second, development of desorption methodology should be performed to assess regeneration time, energy requirements, and potential product concentration and purity.

Silver-based sorbents (AgZ and AgAero) for use in iodine capture from the DOG were evaluated using the established criteria. These sorbents are significantly better understood for this application as a result of Off-Gas Sigma Team efforts during the past decade. The potential implementation of AgAero at a large scale is hindered by its physical degradation by components of the DOG stream, and ongoing efforts to improve the mechanical stability of AgAero should continue.

Much less is known regarding the adsorption of iodine by these sorbents from other off-gas streams in the plant. Iodine management within a UNF reprocessing plant could require treatment of multiple process streams with characteristics differing from the DOG stream (including different iodine speciation, iodine concentration, flowrate, and balance gas composition). Initial efforts have been conducted at both ORNL and INL to understand organic iodine (such as would be found in the VOG) adsorption by AgZ and AgAero, but efforts are currently limited by the level of resources required to address the overarching questions associated with this topic. Testing between these two laboratories on this topic is closely coordinated, and the scientific basis for the testing has been fully developed (Jubin et al. 2017c). Future work should expand this experimental program.

Notably, limited research is available about iodine capture from waste processing operations, including iodine capture from the vitrification off-gas. An analysis of this gas stream should be conducted to better understand this potential application for iodine sorbents.

A review of iodine waste form research and development shows that this area is diverse in nature and that many promising waste forms have been identified for the long-term immobilization of radioactive iodine. Efforts related to the direct conversion of iodine sorbents (including AgZ and AgAero) should be continued because of the potential advantages of direct conversion in a waste management strategy, and other sorbents should continue to be developed and advanced as merited. Development of testing methods that can be used in evaluation of iodine waste forms should continue.

In conclusion, thorough lists of performance criteria and associated metrics have been developed for sorbent and waste form evaluation. These criteria address physical, radiological, and chemical characteristics; technical practicality; technical maturity; cost; and, for sorbents, system performance. The sets of criteria and associated metrics appear to be sufficiently robust and should be applicable whether the wastes containing the four volatile radionuclides (^{3}H, ^{14}C, ^{85}Kr, and ^{129}I) are ultimately classified as low-level or high-level waste. Further, they appear to be sufficient to address both aqueous reprocessing and electrochemical reprocessing of UNF and may be found applicable to advanced reactor off-gas management.

These sets of criteria and associated metrics can serve as tools to evaluate performance at multiple stages within the research and development process. This was exemplified in Revision 1 for technologies relating to krypton/xenon separations and iodine capture from off-gas streams arising from UNF reprocessing. It is expected that the criteria and metrics, as well as the technical evaluation provided in Revision 1, will be beneficial in further developing programmatic direction.

8. ACKNOWLEDGMENTS

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APPENDIX A: DOCUMENTATION OF REVISION 1

This appendix documents the major changes between Rev. 1 and the original document.

- Summary updated to include additions from the balance of the document
- Acronyms and Table of Contents updated to include revisions from balance of document
- Section 1 updated to include focus of Revision 1; additional minor revisions for readability
- Section 3.1 expanded discussion of capacity metric
- Section 3.1 expanded discussion of mechanical stability metric
- Section 3.2 replaces Sections 3.2–3.5 of the original document.
- Section 3.3 removed “Particle Density” as a metric
- Renumbered section 3.6 as section 3.3.
- Section 4.1: Minor updates for readability
- Section 4.3: Minor updates for readability
- Renamed *Section 5: Case Study for Iodine Removal by* $AgNO_3$*-coated Berl saddles and Ag-faujasite* to *Section 5: Implementation Case Study*. Condensed *Section 5: Implementation Case Study* in the main document and transferred unabridged original content to *Appendix B*.
- Addition of *Section 6: Technological Development of* ^{85}*Kr and* ^{129}*I Removal Processes*
- Renumbering of *Section 6: Summary Conclusions* to *Section 7: Summary and Conclusions*.
- Updated *Section 7: Summary and Conclusions* to reflect the findings of Section 6 and the minor changes included in the remainder of the document.
- Added *Section 8: Acknowledgments*
- Renumbered *Section 7: Bibliography* to *Section 9: Bibliography*
- Added *Appendix A: Documentation of Revision 1*
- Added *Appendix B: Case Study for Iodine Removal by* $AgNO_3$*-Coated Berl Saddles and Ag-Faujasite*, containing unabridged Section 5 content from Revision 0

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APPENDIX B: CASE STUDY FOR IODINE REMOVAL BY $A_{G}N_{O3}$-COATED BERL SADDLES AND A_{G}-FAUJASITE

$AgNO_3$-coated Berl saddles were used in the T and B Plants of the Hanford, Washington, facility in the 1950s to limit ^{131}I release to the atmosphere (Cederberg et al. 1961, O'Brien et al. 1963, Paas et al. 1951, McNabney and Lyon 1949). Berl saddles are aluminum oxide (Al_2O_3) support media that served as packing material for the reactive silver component of the adsorbers. Berl saddles were traditionally coated with $AgNO_3$ within the separation facility. Upon column breakthrough, the saddles were processed to remove iodine (as AgI) and were reused with fresh $AgNO_3$ coating.

Silver-exchanged zeolites have been investigated in Europe, Japan, and the United States for their ability to remove iodine from the off-gas streams arising from nuclear fuel reprocessing. There are multiple types of zeolites, primarily distinguishable by their varying Si:Al ratios. Silver-exchanged faujasite, commonly designated AgX, is not currently used in a used nuclear fuel reprocessing facility but has previously been used in both the United States and Italy (Jubin 1988). AgX demonstrates a high capacity for iodine but was eventually discarded in favor of more acid-resistant materials.

Where possible, references for the values or judgements contained within the case study are provided. In some cases, general knowledge is included in the table without citations. In other cases, values were not easily obtained during the course of this study and are designated as unavailable. This does not mean that the metric value has never been measured, but only that our study did not find a reliable source for that particular metric.

Table B-1. Metrics for technical performance and physical and chemical characteristics criterion for selected iodine sorbent comparison.

Property	Unit	Desired trend	$AgNO_3$ coated Berl saddles	Ag-faujasite	Comments
Capacity	mol/m^3	High	12.6	600–2,000[b]	Up to 30% silver utilization in silver saddles; up to 100% in in zeolites (McNabney and Lyon 1949, Katoh 2011).
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	Good	Good	Silver-sorbents highly selective for iodine, with some tramp halogens.
Particle density	kg/m^3	High	n/a	700–1,000	Commercially available sodium form is 673 kg/m^3; density will increase with silver exchange.
Surface area	m^2/g	High	Unavailable	Unavailable	
Specific heat capacity	J/(K·kg) or J/(K·m^3)	Application dependent	See note	Unavailable	Although $AgNO_3$ heat capacity is high, the heat capacity of Berl saddles is low.
Thermal conductivity	W/(m·K)	High	Unavailable	Unavailable	
Radiation stability	% degradation in capacity over time as a function of radiation exposure	High stability; low degradation over time	Very good	Good	$AgNO_3$ refreshed upon saddle column breakthrough; degradation minimal within the time it is expected to remain online. Zeolites are relatively stable in the presence of radiation.
Mechanical stability	Generated fines <420 µm with losses to the off-gas stream of <50 µg/m^3	High stability; low fines generation	Very good, but actual values unavailable	Good[c]	(Puppe and Wilhelm 1990)

Table B-1. Metrics for technical performance and physical and chemical characteristics criterion for selected iodine sorbent comparison (continued).

Property	Unit	Desired Trend	$AgNO_3$ coated Berl saddles	Ag-faujasite	Comments
Thermal stability	% degradation in capacity over time at selected operating temperature	High stability; low degradation over time	High	High	
Chemical stability	% degradation over time as a function of other species present in gas stream	High stability; low degradation over time	High stability	Degrades in acid and high humidity	(Jubin 1981)
Reactivity	Compatibility as determined by standardized compatibility tables	Demonstrated compatibility with all components of gas stream and materials of construction	Compatible	Faujasite may not be acid resistant	(Jubin 1981,Heeb 1994, Cederberg et al. 1961, O'Brien et al. 1963, Paas et al. 1951, McNabney and Lyon 1949)

Table B-2. Metrics for technical practicality criterion for selected iodine sorbent comparison.

Property	Value	Desired trend	$AgNO_3$ coated Berl saddles	Ag-faujasite	Comments
Regeneration	No. of cycles before degrading to 80% of capacity for the target element	High	n/a	n/a	Silver-based sorbents can be regenerated, but it is more likely that they would be disposed of as waste because the iodine is chemisorbed to the silver present in the structure, creating the low-solubility compound AgI.
Co-adsorbed species	mol/kg	Small in number of species and quantity	Low, halogens and antimony as SbH_3 only	Moderate, halogens and ^3HHO	Both Ag-based sorbents will adsorb tramp halogens (e.g., F^-, Cl^-); zeolites will also adsorb ^3H as ^3HHO.
Robustness	% variation in operating parameters tolerated without deleterious effects	High	Medium	Medium	
Flexibility and pretreatment	Operating ranges; no. of unit operations for pretreatment	High flexibility; minimal pretreatment	Some pretreatment required	Some pretreatment required	Saddle columns must be heated to avoid condensation from the process off-gas; faujasite does not tolerate humidity well.
Process complexity	Number and type of control systems and unit operations required	Low	High	Low	For Ag-saddles, upon breakthrough, the column is shut down, sodium thiosulfate is used to remove the AgI, the Ag is either recycled or sent to waste (the usual choice), the saddles are heated and dried, fresh $AgNO_3$ is added to coat the saddles, and the column is dried. No markedly complex operations for Ag-faujasite.
Energy consumption	kW/mol	Low	Unavailable	Unavailable	
Environmental safety and health	Classification according to National Fire Protection Association (NFPA) ratings	Low	Good	Good	Both sorbents have NFPA health ratings of 2 and are considered Resource Conservation and Recovery Act waste.

Table B-3. Metrics for system design and performance criterion for selected iodine sorbent comparison.

Property	**Value**	**Desired trend**	**$AgNO_3$-coated Berl saddles**	**Ag-faujasite**	**Comments**
Pressure drop	Pa/m	Low	Unavailable	Unavailable	Pressure drop for both is expected to be relatively low. Silver-faujasite may have slightly higher pressure drop because of smaller particles and bed packing. Berl saddles were used routinely in the Purex Plant at Hanford (McNabney and Lyon 1949, Cederberg and MacQueen 1961), implying acceptable pressure drops.
Decontamination factor (DF)	$[\text{Isotope}]_a/[\text{Isotope}]_b$ (Unitless)	High	25–200	>1,000	For Berl saddles, reference Cederberg et al. 1961, McNabney and Lyon 1949, and Moore 1984. For Ag faujasite, reference Thomas et al. 1978 and Jubin 1981.
Bed volume	m^3	Low	0.364 m^3 (packed bed)	Unavailable	See McNabney and Lyon 1949 for volume of Berl saddle column.

Table B-4. Metrics for technical maturity criterion for selected iodine sorbent comparison.

Property	Value	Desired trend	$AgNO_3$ -coated Berl saddles	Ag-faujasite	Comments
Technology readiness level of sorbent system	1–9	High	9	6-7	$AgNO_3$-coated ceramic saddles have been used in reprocessing facilities (Wershofen and Aumann 1989). Testing has been done on Ag-faujasite (Thomas et al. 1978).
Commercial availability	Yes/No	Readily available	Yes	No	Silver-saddles are manufactured in place and the precursor materials are available. Sodium faujasite is commercially available.
Time to commercialization	y	Short	None	Very short	Silver-faujasite can easily become commercially available, and all other required materials and unit operations are available and well-developed.

Table B-5. Metrics for cost criterion for selected iodine sorbent comparison.

Property	Value	Desired trend	$AgNO_3$ - coated Berl saddles	Ag-faujasite	Comments
Cost of sorbent material	\$/kg, \$/Ci, or \$/mol	Low	Moderate	Moderate	Both sodium-exchanged faujasite and the saddles (or any other Al_2O_3 support media) are readily available and inexpensive $AgNO_3$ is the cost-driving chemical. ($AgNO_3$ can be used to produce silver-exchanged faujasite, if it is not purchased commercially.)
Operating cost	\$/Ci	Low	Moderate	Low	There are several unit operations that increase the cost of saddle operation: regeneration involves taking the bed offline; washing and dissolving the AgI in a solution of $Na_2S_2O_3$; oxidizing the I^- to I_2; extracting the I_2; reclaiming the Ag; and reloading the support media (Heeb 1994, McNabney and Lyon 1949, Moore 1984, Vignau et al. 1991, Warren 1961). Silver-exchanged faujasite is cost-effective and requires limited unit operations.

The metrics, values, and comments provided in Tables B-1 through B-5 reveal several important trends. First, not all metrics will apply to every sorbent system. An example of this would be the regeneration metric, which is not of interest for either $AgNO_3$-coated Berl saddles or for silver-exchanged faujasite.

Second, if sorbent systems are markedly different, the values provided by the metrics may be disparate on first inspection and should be carefully interpreted in the context of the specific technologies being evaluated. This is illustrated in the capacity metric for the case study, where the capacity (provided in mol/m^3) is notably higher for silver-exchanged faujasite. In this instance, the capacity information should be reviewed in concert with other metrics such as density.

Third, the use of general process knowledge or subjective rankings was often required to complete the case study. This is illustrated by the thermal stability metric, where both technologies were designated "good." Ideally, a more quantitative analysis would be made, but such data were not available. This was a recurring theme in the completion of this case study, and it shows that the performance criteria can be used not only for down-selection between two technologies but also can aid in identifying the knowledge gaps (and their associated importance) that should be resolved during a sorbent or waste form development process.

To conclude the case study, the Berl saddles demonstrated promising technical characteristics in multiple categories that were considered of "high" importance, such as thermal, chemical, and radiation stability. However, the technical practicality metric of process complexity (also of high importance) was scored poorly for Berl saddles. Silver-exchanged faujasite possesses high iodine capacity and is of low process complexity but is less chemically stable than $AgNO_3$-coated Berl saddles. In making a final selection between these two technologies, the factors that are most important for specific plant design should be revisited to complete a determination of the optimal technology.