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Technical Assessment of Off-Gas System Technologies for Potential Use in Molten Salt Reactors

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Executive Summary

Molten salt reactors (MSRs) are a class of Generation IV advanced reactor technologies aimed to enhance and improve the safety, fuel utilization and cost-effectiveness of nuclear power generation. MSR concepts are based on using a molten salt mixture as a primary nuclear reactor coolant, while the fuel can be either directly dissolved in the coolant (i.e., fluid-fueled) or can be in a separate solid form (i.e., salt-cooled solid-fueled). MSRs can be operated in either thermal or fast neutron spectra, as simple fissile convertors, or breeder reactors (e.g., utilizing both fissile and fertile fuels), as well as accelerator-driven sub-critical reactors or transuranic element burners.

Operation of MSRs will necessitate management and treatment of the off-gas streams released to the headspace of the reactor where, in some designs, a cover gas can be circulated to remove certain fission products and maintain an inert atmosphere. An inert MSR cover gas swept over or sparged into the salt will confine radionuclides emerging from the free surface, including radioactive noble and non-noble gases, aerosols, and volatile species. The elements and compounds of these species will have a wide range of thermochemical and physical properties, some of which will decay to different products in the off-gas stream. Therefore, system components used to manage and treat MSR off-gas streams will need to be robust under dynamic reactor and chemistry conditions to support adequate reactor performance by managing the composition and species in the salt coolant and cover gas space. Off-gas system components would perform as a radionuclide boundary to the reactor vessel and a primary means for preventing radionuclide release; therefore, they are of high safety significance for MSR operations.

This report reviews various technologies and materials for potential implementation in the design of an MSR off-gas system per a general conceptual framework. The reviewed technologies operate based on scrubbing, capture, delay, and/or separation of species in the off-gas stream. These include molten hydroxide scrubbers, solid sorbents, delay off-gas systems, particle traps, and cryogenic distillation. Since aspects of an off-gas system will be design-specific, the approach is to provide general discussions on attributes of these technologies and materials, including target off-gas stream species, operational parameters and conditions of importance to decontamination factors, implementation maturity, system component monitoring, potential off-normal conditions, and considerations for waste products. The information aims to support the U.S. Nuclear Regulatory Commission in their safety evaluations for MSR designs. It is noted that this report does not make any conclusions about the conformance of any specific technology or design with regulations that would apply to MSR off-gas systems.

Acronyms and Abbreviations

ALSTS	Axial Flow Large-Scale HEPA Filter Test Stand
ANM	Australian Nuclear Medicine
ARDC	Advanced Reactor Design Criteria
ASME	American Society of Mechanical Engineers
CFR	*Code of Federal Regulations*
d	day
DF	decontamination factor
DOE	Department of Energy
HEPA	high-efficiency particulate absorbing
HIP	hot isostatic pressing
ICET	Mississippi State Institute for Clean Energy Technology
ITS	important to safety
LLNL	Lawrence Livermore National Laboratory
LWR	light-water reactor
MHTGR	modular high-temperature gas-cooled reactor
min	minute
MOF	metal organic framework
MPa	megapascal
MSBR	Molten Salt Breeder Reactor
MSR	molten salt reactor
MSRE	Molten Salt Reactor Experiment
NRC	U.S. Nuclear Regulatory Commission
ORNL	Oak Ridge National Laboratory
PAN	polyacrylonitrile
PDC	principal design criteria
PNNL	Pacific Northwest National Laboratory
RLSTS	Radial Flow Large-Scale HEPA Filter Test Stand
S	second
SFR	sodium-cooled fast reactor

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1.0 Background

Molten salt reactors (MSR) are a class of Generation IV advanced reactor technologies aimed to enhance and improve the safety, fuel utilization, and cost-effectiveness of nuclear power generation. MSR concepts are based on using a molten salt mixture as a primary nuclear reactor coolant, while the fuel can be either directly dissolved in the coolant (i.e., fluid-fueled) or can be in a separate solid form (i.e., salt-cooled solid-fueled). MSRs can be operated in either thermal or fast neutron spectra, as simple fissile convertors, or breeder reactors (e.g., utilizing both fissile and fertile fuels), as well as accelerator-driven sub-critical reactors or transuranic element burners.

Operation of MSRs will necessitate management and treatment of the off-gas streams released to the headspace of the reactor where, in some designs, a cover gas can be circulated to remove certain fission products and maintain an inert atmosphere (Andrews et al. 2021). An inert MSR cover gas swept over or sparged into the salt, will confine radionuclides emerging from the free surface, including radioactive noble and non-noble gases, aerosols, and volatile species. The elements and compounds of these species will have a wide range of thermochemical and physical properties, some of which will decay to different products in the off-gas stream. Therefore, system components used to manage and treat MSR off-gas streams will need to be robust under dynamic reactor and chemistry conditions to support adequate reactor performance by managing the composition and species in the salt coolant and cover gas space. Off-gas system components would perform as a radionuclide boundary to the reactor vessel and a primary means for preventing radionuclide release; therefore, they are of high safety significance for MSR operations.

The U.S. Nuclear Regulatory Commission (NRC) tasked Pacific Northwest National Laboratory (PNNL) with evaluating potential systems and technologies for managing off-gas effluents from MSR designs and identifying safety considerations to system performance. This report provides information regarding systems and technologies based on scrubbing, capture, delay, and/or separation of species in the off-gas streams. The evaluations in this report considered public information and were informed by the authors' engineering experience and judgement per relevant applications. The report was developed in fulfillment of task order 31310022F0033, Task 3, *Technical assessment of off-gas management systems and components for effluents from molten salt reactors*. Consistent with task order 31310022F0033, Task 3, this report only addresses fluid-fueled MSRs. It is recognized that salt-cooled solid-fueled MSRs are likely to generate a lower loading of fission and radionuclide products in the salt coolant due to retention within the pressure boundary of the solid fuel compacts, and therefore a simpler off-gas system in comparison to salt-fueled MSRs.

1.1 Off-gas Composition and System Design

The Department of Energy (DOE) has previously sponsored a report, "Molten Salt Reactor Engineering Study for Off-Gas Management" (McFarlane et al. 2020), which provides extensive information regarding potential off-gas management and treatment technologies for MSRs. Elements discussed in that report include identification of potential components in the off-gas stream, online monitoring of the off-gas stream and system, identification of components of a prototypical MSR off-gas treatment system, normal operations, potential off-normal conditions, and waste management considerations.

The gas stream composition and operational parameters for an off-gas system will depend on the specific MSR design and reactor characteristics (e.g., fuel salt composition, neutron energy spectrum, power level, fuel burnup). The gas stream could also potentially include products generated by degradation (e.g., corrosion) of reactor components from reaction with the coolant salt and radionuclides generated during operation. Kiosidou et al. (2024) have recently reviewed the current state of knowledge on direct and indirect effects of fission products with structural steels under consideration for MSR designs. These products may be mitigated (e.g., via redox control of the salt), and their presence in the off-gas stream would be specific to an MSR design and operations.

Despite these variables, McFarlane et al. (2020) described the main types of constituents that could be entrained in the MSR off-gas as expected to be consistent across the different reactor concepts. These types of constituents include mists, aerosols, particles, volatile species, tritiated products, short-lived volatile radionuclides and daughter products, longer-lived radionuclides, water vapor, non-noble gases, and cover gas, as shown in Table 1-1.

Table 1-1. Molten salt reactor off-gas constituents (modified from McFarlane et al. 2020).

Constituent Type	Examples
Mists, aerosols, and particles	Pyrolysis products,[‡] salt residues, condensed particulates, graphite debris for graphite-moderated fluoride systems, noble metals (e.g., Ru, Pd, Rh), corrosion products (e.g., $CrCl_X$)
Volatile species	Kr, Xe,^{3}HF, HF, H_2O, O_2, F_2, Cl_2, Br_2, I_2, Ar, interhalogens (e.g., ICl, IF_5, IF_7), volatile halides, and the decay products (e.g., Cs, Ba, Rb, Sr, La, Br, I, Se, Te)
Tritiated products	$^{3}H_2$, ^{3}HH, ^{3}HF, ^{3}HF, and possibly ^{3}HHO and/or $^{3}H_2O$
Short-lived volatile radionuclides and their daughters	^{139}Xe $t_{1/2}$ = 39.5 s, ^{90}Kr $t_{1/2}$ = 32.3 s, ^{137}Xe $t_{1/2}$ = 3.83 min, ^{135m}Xe $t_{1/2}$ = 15.3 min, ^{89}Kr $t_{1/2}$ = 3.18 min, ^{135}Xe $t_{1/2}$ = 9.1 h, ^{88}Kr $t_{1/2}$ = 2.84 h, ^{133m}Xe $t_{1/2}$ = 2.19 d, ^{133}Xe $t_{1/2}$ = 5.25 d
Longer-lived radionuclides	^{85}Kr $t_{1/2}$ = 10.7 y, ^{36}Cl $t_{1/2}$ = 3×10^5 y, ^{79}Se $t_{1/2}$ = 6.5×10^4 y, ^{129}I $t_{1/2}$ = 1.57×10^7 y
Non-noble gases	N_2, O_2
Nonradioactive cover gas	He

[‡] MSRE operation showed lubricating oil from the pump bearings entered the pump bowl leading to pyrolysis products in the off-gas (Rosenthal et al. 1972).

McFarlane et al. (2020) have also developed a conceptual framework for an overall MSR off-gas system per prior experience from the Oak Ridge National Laboratory's (ORNL) Molten Salt Reactor Experiment (MSRE), design experience from ORNL's Molten Salt Breeder Reactor (MSBR), and current research activities sponsored by the DOE. This framework identifies various technologies and associated materials that may be included in MSR off-gas system designs and operations for NRC consideration. Therefore, this report has primarily focused on these technologies to discuss performance, maturity, safety considerations for normal operations, anticipated maintenance activities, potential off-normal conditions, and waste considerations. The discussions also address monitoring and control approaches that may be used for successful operation of an off-gas system. Some technologies evaluated in this report have been previously applied in nuclear applications, whereas others are early in the development process.

Figure 1-1 illustrates the overall off-gas system framework developed by McFarlane et al. (2020), which incorporates multiple off-gas systems. A front-end system component, not shown in Figure 1-1, could be a drain hold-up tank configured to provide a 48-hour delay, allowing decay of shorter-lived radionuclides and rejection of decay heat. From that tank, a molten hydroxide scrubber, could be implemented for removal of particles, mists, aerosols, and reactive volatile species from an off-gas stream directly fed from an initial hold-up tank. The working fluid in this system would allow scrubbing of insoluble particles (i.e., removal of reactive species with the working fluid from these particles), as well as chemically dissolving and neutralizing other feed-in gas species. The off-gas stream would then flow through a series of traps for capturing residual halides, water, and oxygen for gas preconditioning prior to downstream delay systems and cryogenic trapping. The delay system, proposed as sorbent beds by McFarlane et al. (2020), could also be a combination of delay tanks or multi-chamber pathways, that would allow further decay of short-lived noble fission gases. A variety of sorbent materials of different technical maturity may be used in the delay system, as discussed in this report. After a suitable decay period, cryogenic distillation, in lieu of sorbent beds, could be implemented to potentially separate stable noble gases from remaining noble fission gases, as well as purification of the carrier gas to be recycled back to the reactor headspace.

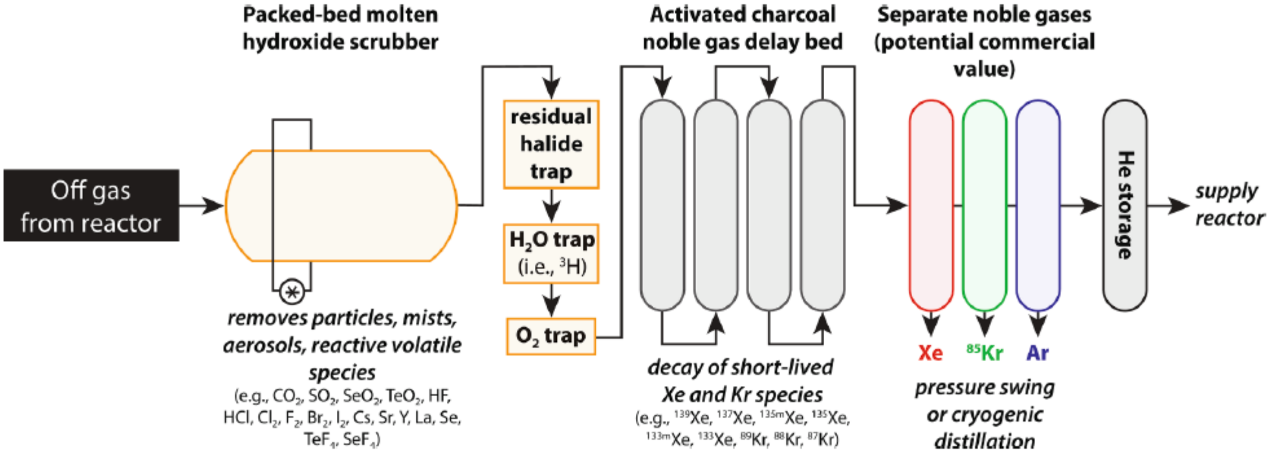


Figure 1-1. Schematic representation of the off-gas system framework for a commercial molten salt reactor based on experience of the Molten Salt Reactor Experiment (Riley et al. 2018; McFarlane et al. 2020).

Per this framework, the off-gas system technologies and materials considered include a molten hydroxide scrubber (Section 3.0), solid sorbents (Section 4.0), delay off-gas systems (Section 5.0), particle traps (Section 6.0), and cryogenic distillation (Section 7.0). It is recognized that the sequence of components and operations for an off-gas system may vary across MSR designs relative to Figure 1-1. The location of each off-gas system component (e.g., the molten hydroxide scrubber in Figure 1-1) will depend on the fuel salt loop design and available surge cover gas volume for the specific MSR design. The available surge cover gas volume in the MSRE was located within the pump bowl, a tank in which the fuel salt pumps were submerged (Rosenthal et al. 1970). However, an MSR design may incorporate a dedicated component at a high point of the fuel salt loop for connection to the off-gas system (McFarlane et al. 2019c). Alternatively, later designs have included the salt drain tank for initial hold up, giving it a dual purpose for radionuclide confinement. Some reactor designs do not have a swept cover gas system. In all of these approaches, it is recognized that the off-gas system is part of the primary containment boundary of the reactor.

1.2 Design Criteria

Regulatory Guide 1.232, “Guidance for Developing Principal Design Criteria for Non-Light Water Reactors” (NRC 2018), describes the NRC’s proposed guidance on how the general design criteria in Appendix A, “General Design Criteria for Nuclear Power Plants,” of Title 10 of the *Code of Federal Regulations* (CFR) Part 50, “Domestic Licensing of Production and Utilization Facilities,” may be adapted for non-light-water reactor (non-LWR) designs. The guidance may be used by non-LWR designers, applicants, and licensees to develop principal design criteria (PDC) for any non-LWR designs, as required by the applicable NRC regulations, for nuclear power plants. This report discusses potential considerations for the design of an MSR off-gas system as informed by Regulatory Guide 1.232 (Section 2.0).

1.3 Decontamination Factors

The decontamination factor (DF) is the primary determinant of the effectiveness of an off-gas system, its components, or associated materials. DF is defined by Equation (1-1), where control efficiency refers to the mass percent of the target analyte captured by the off-gas system or component. For example, a DF of 100 means that 99 percent of the target analyte is captured (Soelberg et al. 2023). The DF of a particular material or technology is expected to be defined as range that depends on various parameters, as listed in Table 1-2. Based on specific reactor designs and the safety analyses for those designs, a DF may help determine the design basis life of a particular off-gas technology. The design-basis life is expected to consider maintenance, waste generation from periodic processing, and/or replacement of contaminated system components, all of which may be needed to meet DF criteria.

$$DF=100/(100\text{-control efficiency}) \qquad \text{Equation (1-1)}$$

Table 1-2. Design and operational parameters impacting decontamination factor range.

Parameter	Notes
Saturation/Fouling/Plugging	The activated surface area of the material or bed will decrease as loading of target species increases, therefore decreasing the measured DF over time. Potential plating of solid particulates along pipes or plugging of filters may decrease flow rates, lead to pressure drops along the system and affect the measured DF.
Degradation	Radiation and/or thermal-induced damage, as well as chemical degradation (e.g., corrosion) of the sorbent or filter material may decrease the measured DF over time.
Off-gas flow rate and/or operating pressure	The residence (hold-up) time and bind/breakthrough of the target species in the sorbent or bed material will vary with process flow rate and temperature and affect the measured DF.
Temperature	Removal of target species by chemisorption and physisorption using activated sorbent materials will depend on operating temperatures.
Volume and surface area	Decontamination will depend on the surface area available for contact between the sorbent or bed material and target species.
Off-gas composition	Decontamination will be affected by the presence of competing species and undesirable impurities. Materials may be designed for selective capture (e.g., Xe over Kr).

Parameter	Notes
Mechanical Failure	Mechanical failure such as a rip in high-efficiency particulate absorbing (HEPA) filter media or a pressure vessel leak could cause an immediate decrease in DF.
Resuspension‡	Resuspension of target species following shutdown activities or during maintenance activities could affect radionuclide transport in the cover gas and off-gas system.

‡ Particle resuspension (or re-entrainment) refers to the removal of particles from surfaces or agitation of the salt.

The design for an MSR off-gas system would define DFs for all reactor operational conditions and system components. Design-specific DFs may need to consider regulatory limits for different species, as currently defined by the pertinent Parts of the CFR; for example, 10 CFR Part 20, 10 CFR Part 50, 10 CFR Part 100 and 40 CFR 190.10, the latter of which stipulates regulatory limits for the release of ^{3}H, ^{14}C, ^{85}Kr, and ^{129}I from the uranium fuel cycle.

The off-gas profile will vary between MSR designs, as will the utility of any specific off-gas system component and achievable DFs. The off-gas stream composition from a reactor facility is expected to be dynamic. Reactor steady-state operation, maintenance activities, shutdown, and transients will result in different cover-gas flow rate, decay heat loading, and off-gas composition, which the system would be expected to accommodate. It is recommended that vendors consider the design and operations of off-gas systems to address potential gaps that could impact reactor design and operations (e.g., low maturity of certain technologies, commercially available materials may not meet DF targets, etc.).

1.4 Normal Operations

Process control instrumentation may be implemented to monitor performance of the components during normal operation and send alert notifications for off-normal conditions. Trending parameters and design range limits will vary by design and operational characteristics of both the MSR core and the off-gas system components. The parameters may include off-gas composition at inlet and outlet streams, volumetric flow rates, temperatures and decay heat rejection, pressures, DF, process media properties (e.g., molten salt fill level), and radiation damage.

The instrumentation design for an off-gas system component will include sensors, gauges, or analytical characterization techniques. Pressure and temperature sensors available to the commercial nuclear industry may need to be redesigned or adapted for monitoring an MSR off-gas. The location of the sensor or probe will be important, as it would determine susceptibility to fouling and corrosion due to off-gas species. Where radiation levels are too high for electronic devices, local shielding around specific devices could be used or the sensors placed in shield boxes or behind shield walls and sensor lines routed to the monitoring points. Instrumentation may need to be routinely tested and calibrated to determine operability.

Various online monitoring techniques may be implemented for characterization of chemical composition and isotopic speciation of the off-gas stream. Combined optical spectroscopy approaches, such as Raman spectroscopy, UV-visible and photoluminescence spectroscopy, and laser induced breakdown spectroscopy have been proposed (McFarlane et al. 2020), although these methods have been tested in laboratory settings with limited dynamic gas compositions. The MSRE fuel salt was sampled regularly through a sampler-enricher system and characterized by chemical analysis after the fact (McFarlane et al. 2020). New MSR designs would likely implement some of these online techniques, although they would need to be qualified for MSR off-gas stream conditions.

Operating procedures may be established to account for routine maintenance of off-gas system subcomponents and minimize the probability of radionuclide releases during these operations. Operations involving major repairs or component replacement would likely be implemented during reactor shutdown, although shutdown may not be needed for lower risk operations such as particle filter replacement or sensor replacement. These operations would be conducted remotely due to the high radioactivity of these components. Switching components will require continued management of the reactor cover gas as decay and fission gases will continue to accumulate. For instance, a secondary delay system may allow for accumulation of these gases and provide passive heat removal of the off-gas during maintenance or shutdown operations (McFarlane et al. 2020).

1.5 Off-Normal Conditions

This report considers potential off-normal conditions for the materials and technologies possibly implemented in MSR off-gas systems. Table 1-3 defines off-normal conditions generically applicable to most off-gas system components. The root cause of a condition may be related to operation of the reactor or operation of the off-gas system itself. It is recognized that the off-gas systems would operate per dynamic off-gas compositions. As an example, during reactor operation, changes in core temperatures may lead to changes in the thermophysical properties (melting point, boiling point, density, and viscosity) of the fuel salt and its fission product contents, which in turn, may affect fission product retention and transport to the gas phase. Off-normal conditions of the cover gas system may challenge plant operations or safety functions such as containment of radionuclides and may need to be addressed in a timely manner. This report particularly focuses on identifying potential off-normal conditions related to the off-gas system itself.

Table 1-3. Off-normal conditions generically applicable to all off-gas materials and system components.

Off-Normal Condition	Off-gas Composition and Off-Gas System Component Impacts
Loss of off-gas system containment	Changes to off-gas composition (cover gas may no longer be inert), increased risk of corrosion (e.g., excess water vapor in the off-gas system), release of accumulated radionuclides to secondary containment.
Overheating	Increase in temperature, decreased decontamination factor (DF), possible pressurization of the off-gas system (e.g., decreased sorption by decay beds), loss of forced off-gas circulation, and composition of down-stream gas not controlled. Overheating of system may impact long term performance of structural components.
Overcooling	Condensing of vaporized salts affecting the flow of the cover gas and entrained material through the off-gas system, decreased DF, pressurization of system components (e.g., solidification of the molten salt media in the primary scrubber) and loss of forced off-gas circulation.
Plugging/fouling	Decreased DF, pressurization of system components, and loss of forced off-gas circulation.

Off-Normal Condition	Off-gas Composition and Off-Gas System Component Impacts
Radiation damage	Potential for radiolytically-induced damage/aging of subcomponents (e.g., solid sorbents in delay beds).
Loss of control/Loss of shutdown capability	Changes to off-gas composition, increased risk of corrosion (e.g., volatile corrosive species—chlorides, fluorides, water—not captured), decreased DF, pressurization of system components, loss of forced off-gas circulation, and composition of down-stream gas not controlled.
Loss of power	Loss of forced off-gas circulation, build-up of radionuclides and neutron poisons in the reactor headspace, pressurization of system components.

In addition, potential off-normal conditions are identified in later discussions in this report, which are specific to each material or technology. Table 1-4 provides the general criteria used to define those conditions that may cause a system component upset, potential options or redundant/mitigative measures, and general timelines for those actions. It is noted that the off-normal conditions discussed in this report may or may not be applicable to a specific MSR design. Without specific design information, the full range of off-normal conditions may be incomplete. These off-normal conditions are defined as causing cascading effects through the complete off-gas system.

Table 1-4. Criteria considered for off-normal conditions related to material or technology operation.

Parameter	Criteria
Off-Normal Condition	Apparent root cause(s) of off-normal condition.
Potential options	Action(s) to put the system into a safe state and return the system to normal operation, including implementation of potential redundant systems and/or mitigative measures (design-based or operational). Actions may not necessarily allow a state that could sustain continued reactor operations.

1.6 Waste Management

Waste management is another important consideration while deciding the design, materials, and operation of off-gas system components. Figure 1-2 shows a flow diagram of how different types of waste streams can be grouped together with several of the other operating waste streams for size reduction, decontamination, and packaging for very long-term storage as Class A, B, and C low-level waste and greater-than-Class-C wastes. This report discusses waste management considerations for the materials and technologies discussed.

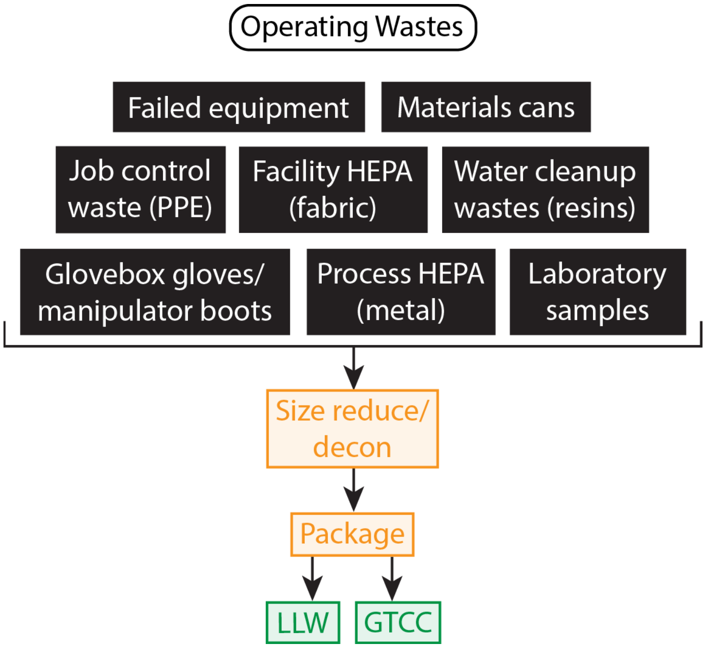


Figure 1-2. Types of reactor operating waste and options for processing (Riley et al. 2018).

2.0 Off-Gas System Design Considerations

Regulatory Guide 1.232, “Guidance for Developing Principal Design Criteria for Non-Light Water Reactors” (NRC 2018), describes the NRC’s proposed guidance on how the general design criteria in Appendix A, “General Design Criteria for Nuclear Power Plants,” of Title 10 of the *Code of Federal Regulations* (10 CFR) Part 50, “Domestic Licensing of Production and Utilization Facilities,” may be adapted for non-light-water reactor (non-LWR) designs. The guidance may be used by non-LWR designers, applicants, and licensees to develop PDC for non-LWR designs, as required by the applicable NRC regulations, for nuclear power plants.

Regulatory Guide 1.232 provides three appendices, which provide design criteria for various non-LWR technologies. Appendix A provides advanced reactor design criteria (ARDC), which are generally applicable to six different types of non-LWR technologies: sodium-cooled fast reactors (SFRs), lead-cooled fast reactors, gas-cooled fast reactors, modular high-temperature gas-cooled reactors (MHTGRs), fluoride high-temperature reactors, and molten salt reactors. Applicants/designers may use the ARDC in Appendix A to develop all or part of the PDC and may choose among the ARDC, or criteria developed for SFRs (Appendix B), or MHTGRs (Appendix C).

Table 2-1 identifies potential considerations for the design of MSR off-gas system components as informed by Regulatory Guide 1.232. These considerations are not meant to be all inclusive, instead are provided as considered relevant per the discussions in this report. It is noted that the NRC will make the final determination on the applicability of these considerations to MSR off-gas system components for a specific design or facility.

Table 2-1. Potential engineering considerations pertinent to molten salt reactor off-gas management and treatment systems.

Design Topic	Potential Engineering Considerations
Fire protection.	The potential of fires and explosions related to the design materials of an off-gas system would be considered. The design of the off-gas system would consider non-combustible and fire-resistant materials wherever practical, and incorporate fire detection and fighting systems, if needed to maintain important to safety (ITS) functions.
Environmental and dynamic effects design bases.	An off-gas system would consider the effects of and compatibility with environmental conditions that may challenge its performance and ability to maintain ITS functions. The off-gas system design would consider dynamic effects that may lead to system failure.
Instrumentation and control.	An off-gas system would include instrumentation to monitor variables related to its safety over an anticipated range of conditions. These variables would consider the relationship of the off-gas system to the MSR core and cover gas containment boundary. The off-gas system would incorporate controls to maintain variables within adequate operating ranges.
Containment design.	An off-gas system would be a radionuclide boundary and associated to the containment boundary. The design of the off-gas system would protect against uncontrolled release of radioactivity and maintain ITS functions during postulated accident conditions.
Electric power systems.	An off-gas system may incorporate electrically powered systems. These systems may have safety functions to ensure radionuclide barriers and other safety functions of the off-gas system are maintained during postulated accidents. These power systems may need sufficient independence, testability, and redundancy.
Containment atmosphere cleanup.	An off-gas system would provide control of fission products and other substances to reduce, consistent with the functioning of other associated systems, the concentration and quality of fission products released to the environment following postulated accidents. The off-gas system would control the concentration of other substances in the containment atmosphere following postulated accidents to maintain containment integrity and related safety functions. Some components of an off-gas system would implement redundancy assuming a single failure.
Inspection of containment atmosphere cleanup systems.	An off-gas system design would permit appropriate periodic inspection of important components.
Testing of containment atmosphere cleanup systems.	An off-gas system used for containment atmosphere cleanup would permit appropriate periodic functional testing of components.
Containment design basis.	An off-gas system would be connected to the MSR core cover gas and containment boundary. The off-gas system would be considered for the containment design basis.
Fracture prevention of containment pressure boundary.	An off-gas system would be connected to the MSR core cover gas and containment boundary. The structural performance of materials used in the off-gas system would be considered for fracture assessment of the containment pressure boundary.

Design Topic	Potential Engineering Considerations
Capability for containment leakage rate testing.	An off-gas system would be connected to the MSR core cover gas and containment boundary. The off-gas system would consider the ability to conduct periodic integrated testing of the containment structure.
Provisions for containment testing and inspection.	An off-gas system would be connected to the MSR core cover gas and containment boundary. Penetrations to the containment structure from the off-gas system would be designed to permit appropriate periodic inspection, with a periodic surveillance program.
Piping systems penetrating containment.	An off-gas system would be connected to the MSR core cover gas and containment boundary. Piping system penetrations to the containment structure from the off-gas system would allow for leak detection, isolation and containment capabilities that reflect the importance to safety of isolating these piping systems. Such piping systems would be designed with the capability to periodically verify, by testing, the operational readiness of any isolation valves and associated apparatus and to confirm that valve leakage is within acceptable limits.
Containment isolation.	An off-gas system would be connected to the MSR core cover gas and containment boundary. Penetrations from the off-gas system to the containment structure would provide for isolation valves, unless demonstrated acceptable otherwise.
Control of releases of radioactive materials to the environment.	An off-gas system would control the release of radioactive gases during MSR operation. Operation of an off-gas system may also generate radioactive solid waste to be managed. The off-gas system would provide sufficient holdup capacity for the retention of gases containing radioactive materials per appropriate environmental considerations.
Fuel storage and handling and radioactivity control.	Operation of an off-gas system may generate radioactive waste to be managed. MSR systems would be designed to maintain adequate safety of such waste.
Monitoring fuel and waste storage.	Operation of an off-gas system may generate radioactive waste to be managed. Appropriate systems would be provided to detect conditions that may result in loss of residual heat removal and excessive radiation levels and to initiate appropriate safety actions.
Monitoring radioactivity releases.	An off-gas system may be connected to the MSR core cover gas and containment boundary. The off-gas system design would provide means for monitoring the MSR containment atmosphere and radioactivity that may be released from operating conditions.
Primary coolant and cover gas purity control	An off-gas system would maintain the composition of the salt coolant and cover gas within specified design limits. These limits may consider chemical attack, potential for fouling or plugging of passages, radionuclide concentration, and air or moisture ingress as a result of a leak of cover gas.
Heating systems	An off-gas system may include heating systems. The designs of these systems and their controls would consider appropriate temperature distributions and rate of change in temperature if a failure occurs. The potential for plugging of a cover gas line would be considered in the design and controls.

3.0 Molten Hydroxide Scrubber System

3.1 Technology Description

A caustic scrubber system is a multi-purpose separation technology designed to remove particulates, mists, aerosols, and reactive volatile gases from an off-gas stream. Scrubbers can be designed either with an aqueous hydroxide mixture or a “nonaqueous” molten hydroxide eutectic mixture. For MSRs, a “nonaqueous” molten mixture may be preferable due to limited presence of water in the system and higher temperatures from MSR off-gas streams. This design would be expected to have a higher efficiency (i.e., DFs) in comparison to an aqueous-based design from the use of a counterflow scrubber. Note that even nonaqueous hydroxides will contain residual levels of H_2O and carbonate species.

A molten hydroxide scrubber would use a eutectic mixture of alkaline hydroxides (e.g., NaOH-KOH-LiOH) with low melting point (T_m) but high boiling points (T_b) (e.g., a 2Na/2K/Li mixture will have T_m ~ 200°C and T_b ~1300°C) to neutralize volatile species and reactive gases and capture particulates and aerosols (McFarlane et al. 2020). The viscosities and densities of these hydroxides are similar to those of aqueous solutions at operating temperatures between 150 and 300°C, although these data are scarce. As described in Riley et al. (2018), the molten hydroxide scrubber is envisioned as a “slow flowing waterfall of a molten hydroxide mixture over a stationary high surface area substrate (e.g., a packed bed of nickel spheres).” Such a configuration will provide for the molten hydroxide to flow down through the bed while the MSR off-gas counterflows up through the bed and out of the top of the scrubber as “cleaner” gas. The design would operate per an adequate range of off-gas counterflow rates (gas velocities) that would prevent or mitigate potential aerosolization of the molten hydroxide salt during the scrubbing process.

The hydroxide scrubber mixture is continuously heated and circulated by a pumping system. It is important that the MSR off-gas system design allow for sufficient gas velocity to prevent solids collection within the off-gas piping upstream of the scrubber. This configuration would behave as a non-pluggable filter, and consideration would be given to periodic replacement and flushing of the hydroxide salt. Fundamentally, hydroxide interaction with the off-gas enables three mechanisms for off-gas cleaning: (1) capture of insoluble particles, (2) chemical conversion to hydroxide salt soluble species, and (3) neutralization of acid species. For example, insoluble metal particulates will be scrubbed or trapped within the hydroxide scrubbing solution, chemically soluble species (MSR salts) will go into solution, and acidic species (e.g., CO_2, SO_2, HCl, HF, SeO_2, and TeO_2) will be neutralized and trapped in solution. Non-condensable gases will pass through the scrubber (e.g., O_2, Ar, He, Kr, and Xe). Conceptually there are other off-gas constituents that could be captured by the molten hydroxide scrubber system (e.g., tritium, Cl_2, F_2, Br_2, I_2, Cs, Sr, Y, La, Se, TeF_4, and SeF_4) (Riley et al. 2018, 2019; Trowbridge et al. 2003); however, further process evaluation is necessary. Fluorine may be generated during processing due to radiolysis of salt residues, which may produce HF or F_2. Further, in the reactive process, such as the formation of iodate from iodine, H_2O is generated. Hence, steam would need to be trapped downstream of the scrubber.

In 2003, a molten hydroxide scrubber was tested to clean radioiodine from off-gas. In this study, iodine was dissolved in the molten hydroxide, converted to an iodide, then cooled. Once cooled, the iodide was separated through melt crystallization. This process had a 96 percent scrubbing efficiency, but hydroxide was found in iodide crystals (Trowbridge et al. 2003; Riley et al. 2018).

In 2019, ORNL reported on recent testing of a packed-bed molten hydroxide scrubber for neutralizing acidic gases. ORNL built a scrubber that uses a mixture of NaOH and KOH and includes a feature to direct particulates and aerosols to a sump for accumulation and removal. The configuration has the gas enter the scrubber through a side port and exit through the top. (Figure 3-1 and Figure 3-2) (McFarlane et al. 2019a).

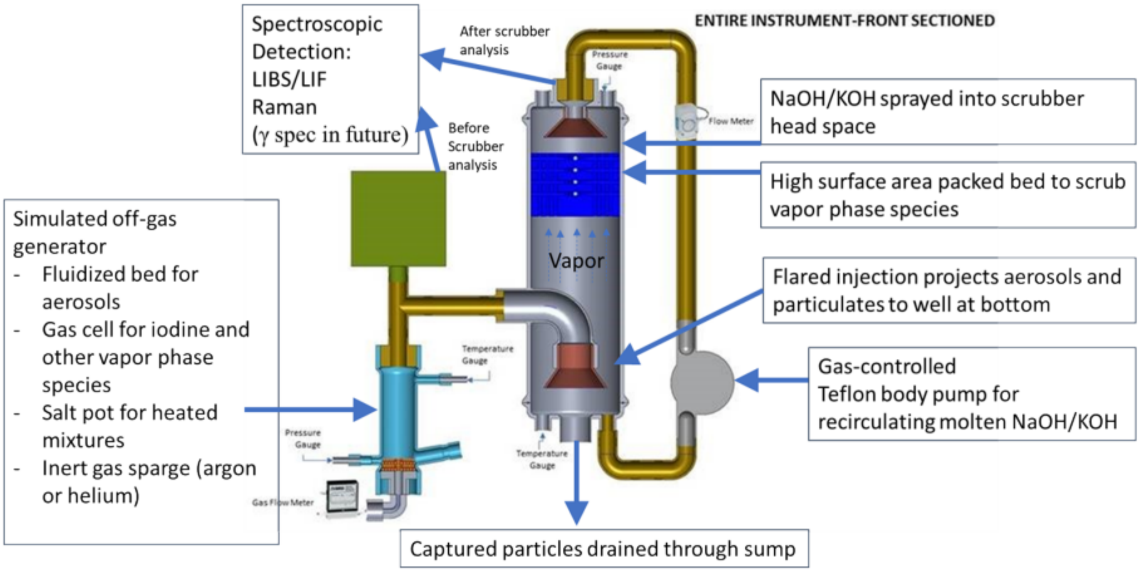


Figure 3-1. Example molten hydroxide scrubber configuration (McFarlane et al. 2019a).

Figure 3-2. Image of the molten hydroxide scrubber prototype system at ORNL. Image courtesy of Joanna McFarlane (ORNL).

3.2 Safety Considerations

3.2.1 Normal Operations

Performance monitoring of the molten hydroxide scrubber is necessary to ascertain that: (1) the bed is functioning properly to achieve target DFs (e.g., as monitored through sensors and/or *in situ* chemical quantification), (2) the temperature of the bed remains above the melting temperature of the hydroxide mixture (below which point it will freeze), and (3) concentrations in the feed-in gas stream, dissolved compounds, and particulate-type species suspended within the scrubber remain within design targets, above which point, the bed may operate under reduced efficiency.

On-Line Process Monitoring

The instrumentation design of a molten hydroxide scrubber system will need to consider a combination of process monitors or sensors for temperatures, pressures, corrosion/erosion monitoring, flow, and radiation monitoring. This will support safe system operations and monitoring of system performance (e.g., DFs). On-line analytical sensors/monitors for trending performance of the hydroxide scrubber bed would be implemented.

Table 3-1 proposes the type of instrumentation for monitoring of the molten hydroxide scrubber, as well as the instrumentation's proposed location and purpose. Instrument and control system design includes at least three possible purposes for various instruments: (1) alarm functions that enable operator alerts if a sensor detects a process condition out of normal range, (2) control functions that enable equipment interlocks or close loop process control, and (3) acquisition functions that enable data collection using in trending system performance. The monitors that provide acquisition functions would include analytical equipment such as Raman spectroscopy, UV-visible, and photoluminescence spectroscopy. Table 3-2 identifies potential species to be monitored by each monitoring technique (McFarlane et. al. 2020). Further development may be needed for the online analytical equipment proposed to monitor off-gas chemistry and any key indicators of the molten hydroxide scrubber bed. These monitors would be important to reducing risks to workers, the public, and the environment, as well as improving the operational availability of the MSR.

Table 3-1. Instrumentation of the primary off-gas system.

Instrumentation Type	Device Location(s)	Purpose (Alarm, Control, Acquisition)	Notes
Temperature: Process off-gas	Upstream and downstream of each major component of the off-gas system.	Alarms for process conditions out of normal ranges.	-
Temperature: Hydroxide scrubber bed	As required to monitor scrubber process conditions.	Acquisition: Trend system performance.	Multiple process thermocouples would be considered.
Pressure: Molten salt reactor (MSR) headspace	Piping between MSR headspace and scrubber.	Control of MSR headspace pressure.	See below for a system pressure control strategy.

Instrumentation Type	Device Location(s)	Purpose (Alarm, Control, Acquisition)	Notes
Pressure: Process off-gas	Upstream and downstream of each major component of the off-gas system.	Acquisition: Trend pressure drop across off-gas system components. Alarm to high pressure conditions in the system or high pressure drop across scrubber and absorber beds.	-
Level: Hydroxide level of the scrubber	Hydroxide scrubber	Alarms to high and low hydroxide level conditions.	-
Flow Rate: Process off-gas	Downstream of last off-gas treatment component.	Acquisition: Trend system performance. Alarm to process conditions out of range of expected flow conditions.	Thermal and Coriolis type mass flow meters.
Erosion/corrosion	Sensors placed at elbows and low points of the piping system to trend loss if pipe wall thicknesses.	Acquisition: Trend erosion and corrosion of the piping system	Installed ultrasonic sensors are a possible option.
Radiation monitors	Sensors placed at elbows and low points of the piping system.	Acquisition: Trend increased source terms to remain informed about material settling in the piping system.	-
Realtime analytical monitors	Off-gas scrubber and piping. Other locations to be determined.	Acquisition: Trend system performance.	Optical spectroscopy techniques (see Table 3-2)
Hydroxide turbidity	Hydroxide scrubber	Alarm on opacity/turbidity	Optical method
Failure of hydroxide containment	Scrubber well	Temperature, conductivity alarm	-

Table 3-2. Potential optical spectroscopic techniques for real-time monitoring of off-gas species (McFarlane et al. 2020).

Monitoring Technique	Potential Species
Raman spectroscopy	HF, HI, H_2O, H_2, $^3H_2$, ^3HH, ^3HF, N_2, O_2, F_2, Br_2, Cl_2, ICl, IBr, BrCl, I_2
UV-vis spectroscopy	I_2, IHI, HBr, Br_2, BrCl, CH_3I, ICl
Photoluminescence spectroscopy	I_2, Br_2, Cl_2, HOI, ICl

As the concentrations of fission products and other impurities reach levels that are high enough to limit performance, it is likely that the hydroxide mixture will have to be replaced to maintain efficiency. Radiolysis of the hydroxide salt may also degrade performance and allow some fission products to re-volatilize. The lifetime of the hydroxide salt and need for replacement and flushing, as well as the operations involved (online processing or diversion of off-gas to a secondary scrubber while the primary scrubber is processed), would likely need additional testing and engineering design. The main purpose of analytical monitors would be to monitor efficiency (e.g., DFs) of the bed and prevent volatiles from passing through; perhaps this could be done with concentration monitors on the downstream side of the bed for key radionuclides requiring capture by a bed of this type. Optical techniques could be evaluated for monitoring the hydroxide salt turbidity as means to assess the need for replacement.

The selection of the materials for scrubber containment would consider corrosivity of the hydroxide salt at operating temperatures (e.g., selection of corrosion-resistant nickel-based alloys). Containment failure could potentially be monitored via conductivity measurements and/or temperature. Further, the pumping system may need to be monitored due to potential radiation damage to subcomponents (e.g., polytetrafluoroethylene seals). The instrumentation needs identified in Table 3-1 for monitoring normal safe operations, all or in part, may vary depending on the system design. The support systems such as general plant systems and molten hydroxide recirculation are not included here. Further consideration, perhaps identified through accident/hazards analysis, would be given to the need for additional instrumentation and redundancy to improve systems reliability and reduce hazard risks. Expected system upsets that would be discussed during accident/hazards analysis would include high/low temperatures, high/low pressures, failure of system exhausters, failure of molten hydroxide recirculation system, and sudden high pressure drops across each of the major off-gas treatment components. Consideration would be given to the need for bypassing to a redundant secondary scrubber system to address an off-normal condition of the primary system or potential failure.

System Pressure Control

Classical means of pressure control of process off-gas systems include using variable speed exhausters, control dampers, and ballast gas control at exhauster inlets. These are well established industry methods; however, they are deployed at the end of the off-gas system train and sometimes cannot react fast enough to upstream process flow variations. Improved pressure control is possible by controlled injection of gas into the MSR headspace or downstream piping. This is accomplished with an automated control loop where gas flow injection is adjusted to maintain system pressure within required ranges. This control feature provides faster responses to variable process conditions which impact system pressures by maintaining steady flows in the off-gas system. This strategy has successfully been deployed in waste vitrification systems including research scale systems at PNNL. Key benefits of controlled gas injection near the headspace include:

- Gas flow can be increased sufficiently to improve gas velocity in the off-gas pipe, reducing solids collection within the off-gas piping.
- Improved process control by decreasing the system response time to variations in off-gas system flow.

3.2.2 Off-Normal Conditions

Table 3-2 provides a partial set of potential off-normal conditions and potential options to address these conditions. A hazards analysis for the proposed scrubber system would identify consequences and risks for potential off-normal conditions of the specific design. The main safety concern with the concept discussed in Section 3.1 is that the molten hydroxide scrubber pre-filtering aspect of the design (see Figure 1-1) requires that it be heated to remain in the molten state. In a loss-of-power condition, the molten hydroxide would freeze into a solid state and a high-pressure condition could arise on the front-end coming from the reactor purge from the core to the off-gas treatment arm of the facility. Potential options center on switching to back-up electrical power, switching to a completely redundant off-gas system, having a redundant control system, and installing redundancy for any critical instrumentation. Timelines would be design specific. It is recommended that consideration be given to the retention of fission products within the solidified hydroxide and downstream changes to the off-gas composition when the hydroxide is remelted.

Table 3-3. Summary of potential off-normal conditions and potential options for the molten hydroxide scrubber technology.

Off-Normal Condition	Potential Options
Loss of heater power and potential freezing of the hydroxide salt and over-pressurization of the off-gas system	Switch power source, start generators, switch to redundant scrubber or backup off-gas system.
System is saturated and no longer working as intended resulting in lower DFs	Switch to a backup system such as a passive solid sorbent bed or redundant hydroxide scrubber.
System breach due to corrosion of confinement components or mechanical stress resulting in lower DFs	Switch to a backup system such as passive solid sorbent bed or redundant hydroxide scrubber.
Failure of instrumentation or control	Switch to redundant system.
Piping plugging or fouling due to hydroxide salt aerosol transport and buildup, leading to high pressure drops downstream	Switch to a backup system such as a passive solid sorbent bed or redundant hydroxide scrubber.
Loss of hydroxide recirculation resulting in lower DFs	Depends on mode of failure. Switch to a redundant recirculation system or backup off-gas system.

3.3 Considerations on Waste Products

It is presumed that the entire hydroxide salt would have to be treated as a waste stream upon removal or replacement of the scrubber bed. It is plausible that specific chemical components could be separated and separately managed; however, the most straightforward process would be a direct immobilization of the salt. A number of waste forms could be considered such as minerals or glass depending on the approach for very long term storage (see also McFarlane et al. 2020: Section 8.2). A conceptual approach would be to precipitate the halides as silver halide and embed in a low melting metallic matrix. Per this approach, the cations and other retained particulates would be immobilized in a borosilicate glass, and the hydroxide with trace radionuclides including tritium would be cemented in high integrity containers. Waste treatment optimization options are likely to reduce the complexity and cost of treatment, storage, transportation, and very long term storage.

3.4 Technology Status

3.4.1 Prior Experience

Aqueous hydroxide (caustic) scrubbers have been implemented at industrial and nuclear facilities (Riley et al. 2016). However, the concept of a molten hydroxide scrubber has only been studied in a laboratory scale. Technology development efforts have been performed to demonstrate the proof of principle (McFarlane et al. 2019b; Trowbridge et al. 2003). Recent work conducted at PNNL focused on monitoring iodine and carbonate species in a molten NaOH-KOH molten hydroxide scrubber bed (Medina et al. 2022). A prototype molten hydroxide scrubber has been built at ORNL and is undergoing testing to evaluate this device for practical implementation. The flexibility of this device to accept different salt compositions, which includes a variety of different eutectic compositions of alkali-hydroxides, operation at different temperatures, and the flexibility of containment materials available provide benefits over aqueous caustic scrubbers. The molten hydroxide scrubber would operate at temperatures well above aqueous caustic scrubbers with similar process features (e.g., particulate capture, pH neutralizations).

3.4.2 Information Gaps or Needs

To the authors' collective knowledge, a molten hydroxide scrubber has not yet been demonstrated at a large scale nor within a nuclear facility. Further testing is needed to advance the technology maturity. The research-scale molten hydroxide scrubber developed and tested by ORNL has proven at least at a conceptual level the design could reasonably be expected to support MSR operations in the future (McFarlane et al. 2020). However, materials performance (e.g., radiation hardness of the hydroxide salt) and scale up questions remain. Design evolution to a full-scale, mature scrubber may require multiple iterations with further research and development. The following are considerations to support potential implementation of a molten hydroxide scrubber for MSR off-gas management.

- Perform a preliminary hazards analysis and technology assessment, possibly before beginning conceptual design. This will identify the technology gaps and produce a more robust set of functions and requirements to advance the scrubber design.
- Further testing and 3-D computational fluid dynamics modeling could be required to understand design requirements before scaling up.
- Establish key performance metrics used for future sizing including off-gas residence times and required operating ranges of the scrubber. Include a chemical analysis for capture of reactive species such as radioiodine.
- Establish engineering scaling methods used to support future sizing calculations.
- Determine the effect of radiolysis on hydroxide scrubber performance and resuspension of captured radionuclides.
- Assess materials that can withstand the hydroxide and the connections between these materials and the upstream process lines that need to withstand exposure to acidic gases.
- Determine if gas to hydroxide separation in scrubber headspace is important to minimize hydroxide downstream carryover.
- Determine if there is an optimal design to prevent backflow of hydroxide salt from entering the MSR cover gas region.
- Determine if there is an optimal bed diameter to height ratio to assess impacts to scrubber performance (e.g., DFs).
- Identify design features critical to reduce pressure fluctuations at the MSR headspace.

- Identify the process for filling and extracting molten hydroxide and the design features required for these processes.

- Implement a molten hydroxide scrubber within an integrated MSR off-gas system to understand system dynamics and develop operating procedures and reduce risks.

- Qualify online monitoring and instrumentation methods per dynamic MSR off-gas compositions and operational conditions (including high-radiation environment) to define sampling and characterization needs to achieve target DFs.

4.0 Solid Sorbents

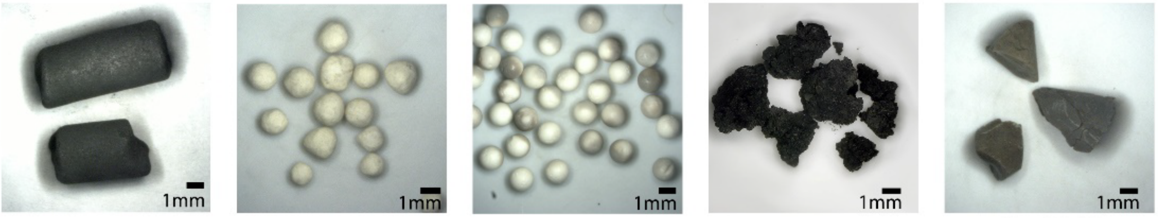
4.1 Technology Description

The category of solid sorbents includes several different types of materials that can be implemented at various stages within MSR off-gas systems to mitigate releases of different species including halogens, noble gases, carbon oxides, and water (see the residual halide trap, H_2O trap, and O_2 trap in Figure 1-1) (Banerjee et al. 2015; Vienna et al. 2015; Riley et al. 2016; Riley et al. 2019). Solid sorbent materials operate through physisorption, chemisorption, or a mixture thereof to bind target species; an alternative method is a molecular sieve-type approach where molecules too large to pass through the pores of the sorbent (engineered through crystal chemistry) can get trapped due to steric constraints.

Solid sorbents include desiccants for pulling out water from the stream (water can lead to corrosion of reactor components), removal of tritiated species (^3HHO or ^3H_2O), activated/functionalized sorbents (e.g., metal-exchanged zeolites, metal-loaded aerogels or xerogels, porous silica or alumina, and activated carbon) for targeting halogens, molecular sieve-type capture media for capturing and separating noble gases from gas mixtures (e.g., metal-organic frameworks, zeolites), and physisorption-based sorbents (e.g., activated carbon).

Mordenite and faujasite are naturally occurring mineral zeolites, although synthetic analogues are used for their controlled pore structure and ability to moderately vary SiO_2/Al_2O_3 molar ratios. Oxide-based aerogels and xerogels are created through hydrolysis and polycondensation reactions, solvent exchange, and drying to achieve a low-density solid form with high specific surface area. The differences between aerogels and xerogels have to do with the physical properties, where aerogels tend to have relatively higher porosities, lower densities, and lower mechanical integrity. Typically, the base processes and materials are the same for aerogels and xerogels, but the difference is the drying method where critical point drying is used for aerogels (i.e., replacing the solvent in the matrix with a supercritical fluid) and xerogels can be produced through ambient pressure drying or drying under vacuum. Porous alumina and porous silica with tailored pore structures and properties are routinely produced with sol-gel methods and template-assisted synthesis. While aerogels tend to have higher specific surface area, xerogels often have more mechanical integrity. Metal organic frameworks (MOFs) are a newer class of high-surface area crystalline porous materials with structures composed of metal cations and organic linkers. MOFs can be designed to adsorb specific molecules, such as using electron-donor functional groups to bind iodine molecules which are electron accepting (Zhang et al. 2022). Activated carbons refer to charcoal that has been treated to increase porosity, and which may be impregnated with compounds such as sulfur, potassium iodide or 1,4-diazabicyclo[2.2.2]octane (DABCO), all of which enhance iodine reactivity and capture. Figure 4-1 and Figure 4-2 illustrate example sorbents for iodine and noble gas capture, respectively.

Solid sorbent materials for excess water removal may be implemented in the MSR off-gas system (see Figure 1-1). As mentioned earlier, the trapping of acidic species such as HCl, HF, or HI in the hydroxide melt of the primary scrubber will form water, which may be monitored downstream by conductivity measurements or Raman spectroscopy (McFarlane et al. 2020). Most of the water products, including tritiated species ^3HHO or ^3H_2O, evolving from the scrubber would be condensed and drained. Any residual water could be removed by downstream water traps, which may incorporate sorbents such as zeolites, aluminosilicate molecular sieves or mesoporous silica gels. As the lower-bound design DF limit is reached, these traps would likely need replacement or less likely, potentially regeneration by heating and off-gassing captured water to a decay tank. Silica gel has higher reversible water adsorption capacity at lower temperatures (if regeneration is considered in the process) relative to zeolites.



BAT37 (AC) **IONEX** **AC6120** **Ag-FA** **Ag-HTX**

Figure 4-1. Iodine sorbent materials: BAT37 activated carbon, IONEX Ag-400 (Ag-faujasite), Clariant AC-6120, Ag-functionalized silica aerogel (Ag-FA), and Ag-functionalized heat-treated aluminosilicate xerogel (Ag-HTX) (modified from Chong et al. 2024).

SBMOF-1 **HKUST-1**

Figure 4-2. Engineered metal-organic framework polyacrylonitrile (PAN) composite beads. Left image: 90 mass% SBMOF-1:10 mass% PAN (Robinson et al. 2024). Right image: 75 mass% HKUST-1: 25 mass% PAN (Riley et al. 2020).

Solid sorbent materials may also be implemented for iodine or noble gas capture in the form of packed beds. As shown in Figure 4-3, some solid sorbent materials show areas of overlap between iodine and noble gas capture. Table 4-1 provides a summary of solid sorbent materials for water, iodine, and noble gas capture, which identifies their maturity for potential implementation in MSR off-gas systems. The sorbents vary between developmental and conceptual options of low maturity (e.g., Ag-aerogel, Ag-xerogel, chalcogels) to options with high maturity, proven by implementation at nuclear facilities (e.g., Ag-mordenite, Clariant AC-6120, activated carbons) (Riley et al. 2016). The materials range in commercial availability and cost.

In this report, sorbents containing Ag use notation such as Ag-mordenite. The exact chemical form of the Ag is not always known for Ag-containing sorbents. However, generally speaking, Ag-containing sorbents behave similarly when reacting with iodine, regardless of the Ag form (i.e., Ag^+ vs Ag^0, present in different compounds like $AgNO_3$), although performances may vary. Based on studies at PNNL (Riley et al. 2017a, Riley et al. 2022), this is the case whether Ag is present as Ag^+ ions (such as sitting in cages/channels in zeolites, charge balancing negatively charged Al-Si-O frameworks), as Ag-salts (Clariant AC-6120 is $AgNO_3$-loaded porous alumina), or as Ag^0 nanoparticles because AgI is formed in all these cases. In all these cases, Ag is the “getter” for binding iodine, and the gettering process is (at least mostly) considered chemisorption, where a new compound is created even if a redox reaction occurs during the process.

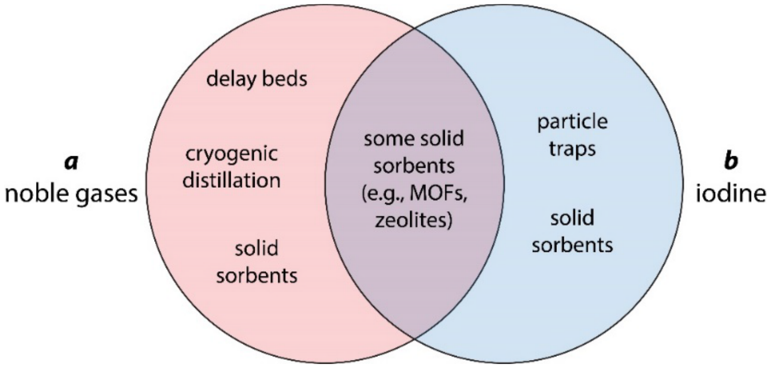


Figure 4-3. Venn diagram showing areas of overlap between (a) noble gas, and (b) iodine capture technologies, where some common sorbents can be used (in different capacities) for both types of streams.

An MSR off-gas system may implement a combination of sorbent materials or sorbent bed sequences, per reactor-specific needs (e.g., target DFs). As noted in Table 4-1, implementation maturity will be an important consideration for demonstrating a particular sorbent material can be fabricated and properly qualified. Some sorbent materials are commercially available and may be sourced from vendors with a Quality Assurance Program consistent with 10 CFR Part 50. Sorbent materials of low technical maturity are not commercially available but are being tested in laboratory scales.

Solid sorbents have designated practical operating conditions per manufacturer specifications, such as: (1) active temperatures, (2) flow rates (off-gas residence/hold-up times), (3) concentrations of oxidizing and undesirable species relative to target species in the feed-in off-gas stream (i.e., need for gas preconditioning), and competing species environments (e.g., capturing $I_{2[g]}$ in the presence of $F_{2[g]}$, $Cl_{2[g]}$, and/or $Br_{2[g]}$). The DF for a sorbent material varies based on the dynamic nature of the MSR off-gas stream and operational conditions, as well as the radiation hardness of the material.

For many or nearly all noble gas capture sorbents, lower-than-room temperature operation is desirable to improve DFs and capacities. The operating temperature range is defined to ascertain that the sorbent material behaves as intended before thermal oxidation (in the presence of $H_2O_{[g]}$ or $O_{2[g]}$), structural changes (e.g., amorphization), or chemical decomposition. Issues arise with solid sorbent performance in the presence of acidic vapors or oxidizing gases, which include reduced capture performance and mechanical degradation of the sorbent. In some cases, the solid sorbent bed can be regenerated and reused. As shown in Figure 1-1, residual halide traps could be used to capture any halides that get through the primary off-gas system for solid scrubbing (e.g., the molten hydroxide scrubber) prior to a sorbent bed.
Table 4-2 provides an overview of some key attributes for consideration for the different sorbent classes. It is noted that sorbent performance (DF and capacity) will depend on the dynamic/radiological conditions of the off-gas stream for a particular MSR design.

Table 4-1. Examples of solid sorbents for different off-gas species.

Off-Gas Stream	Solid Sorbent Type	Example(s)	Implementation Maturity	Reference(s)
Water	Zeolites/ molecular sieves	Y-type zeolites (***commercial***); Mordenite, (***commercial***); 3A molecular sieve (***commercial***)	High	Tashiro et al. (2004); Vienna et al. (2015)
	Silica gel	CARiACT Q Series (***commercial***)	High	IAEA (2004); Matsuoka et al. (2003)
	Desiccant	$CaSO_4$ (***commercial***)	High	Vienna et al. (2015)
Iodine	Zeolites/ molecular sieves	Ag-mordenite (***commercial***), Ag-faujasite; Bi-mordenite	High	Jubin (1981); Yadav et al. (2023); Hao et al. (2023)
	Aerogels or xerogels	Ag-functionalized silica aerogel, Ag-aluminosilicate aerogels/xerogels, chalcogels (sulfide aerogels)	Low	Matyáš et al. (2018); Riley et al. (2017a); Riley et al. (2022)
	Porous silica	Ag-KTB and Ag-KTC (Ag-loaded silicic acid) (***commercial***)	High	Holladay (1979)
	Porous alumina	Clariant AC-6120 (Ag-impregnated)(***commercial***)	High	Wilhelm and Schuettelkopf (1970)
	Carbon	Activated carbon (e.g., S-impregnated Kombisorb® BAT-37 (***commercial***), 1,4-diazabicyclo[2.2.2]octane (DABCO)-impregnated, KI-impregnated or triethylenediamine (TEDA)- (***commercial***)	High	Fujii et al. (2022); Zhou et al. (2014); Huve et al. (2018)
Noble gases	Zeolites	Ag-mordenite (Xe over Kr), H-mordenite (Kr over Xe)	Low	Greenhalgh et al. (2014); Garn et al. (2015)
	Molecular sieves	Metal-organic framework (MOF, e.g., HKUST-1, SBMOF-1)(Xe over Kr)	Low	Robinson et al. (2024); Riley et al. (2020); Zhang et al. (2022)
	Carbon	NUCON® NUSORB® Grade GXK™ (***commercial***)	High	NUCON (2019); Thallapally et al. (2022)

Table 4-2. Attributes of solid sorbents for iodine and noble gases.

Active Species	Solid Sorbent Type	Active (Operating) Temperature	Target Species Capacity (per sorbent mass)	Additional Considerations	Commercial availability
Iodine	Zeolite	100–150°C	>100 mg/g	High thermal and mechanical stability (strength); Capture competition with other halogens	Yes
	Aerogels	100–150°C	>300 mg/g	High chemical stability; Low to Moderate mechanical stability (low density); Good chemisorption, some physisorption; Capture competition with other halogens	No
	Xerogels	100–150°C	>300 mg/g	Good chemisorption; Moderate mechanical stability; Capture competition with other halogens	No
	Porous silica	100–150°C	>100 mg/g	Unknown capture competition with other halogens	Yes
	Porous alumina	100–150°C	>100 mg/g	Capture competition with other halogens	Yes
	Activated carbons	Room temperature or lower	>100 mg/g	Physisorption-only capture can result in desorption of target analyte downstream	Yes
Noble gases	Zeolites	Below room temperature	>30 mmol/kg	Require engineered form (demonstrated in polyacrylonitrile [PAN] composite form)	Yes (precursor), No (engineered form)
	Molecular sieves/MOFs	Below room temperature	>30 mmol/kg	Require engineered form (demonstrated in PAN and polymethyl methacrylate [PMMA] composite form)	Yes (precursor), No (engineered form)
	Carbon	Room temperature or lower	>30 mmol/kg	High thermal and mechanical stability (strength); Qualified for use at nuclear facilities	Yes

Solid sorbent beds would require changeout as DFs are reduced after usage. A sorbent bed lifetime, cost of implementation, DF and total target analyte loading capacity can vary widely across the candidate options. For iodine solid sorbents, the implementation will likely be a packed bed of the solid sorbent in a columnar geometry (Bruffey et al. 2019; Shen et al. 2024). These could be implemented in a parallel setup where a secondary bed is used during change-out of a primary bed after it is loaded (e.g., lead-lag or lead-lag-polish column configuration). An example layout is shown in Figure 4-4 and a schematic of the layout is shown in Figure 4-5. Sorbent beds for both noble gas and iodine capture would require temperature control and pre-scrubbing filtration of fines, particulates, and competing gases to prevent disrupting bed capture operations.

At the Hanford Facility in Washington State, the Waste Immobilization and Treatment Plant is being built to process and stabilize radioactive waste that is currently stored in underground tanks. The plant's High-Level Waste Vitrification Facility has been designed with a primary and secondary off-gas system associated with each high-level waste melter. Two silver mordenite columns are included in the secondary off-gas system design to control the release of ^{129}I and other gaseous halogens (e.g., fluorine, chlorine) (Scheele and Wend 2015; Christie and Miller 2019). The columns will consist of 36 adsorber cartridges filled with silver mordenite in the form of cylindrical pellets. Figure 4-6 provides a design diagram for the high-level waste facility vitrification system. As it is anticipated that the adsorbers will lose effectiveness over time and will need to be replaced, the columns have been designed as a bank of cartridges, which allows for replacement (Ecology 2003).

Figure 4-4. Picture of solid sorbent testing column at Oak Ridge National Laboratory for evaluating capture of long-chain organic iodides on silver mordenite (Ag^0Z) (Bruffey et al. 2019).

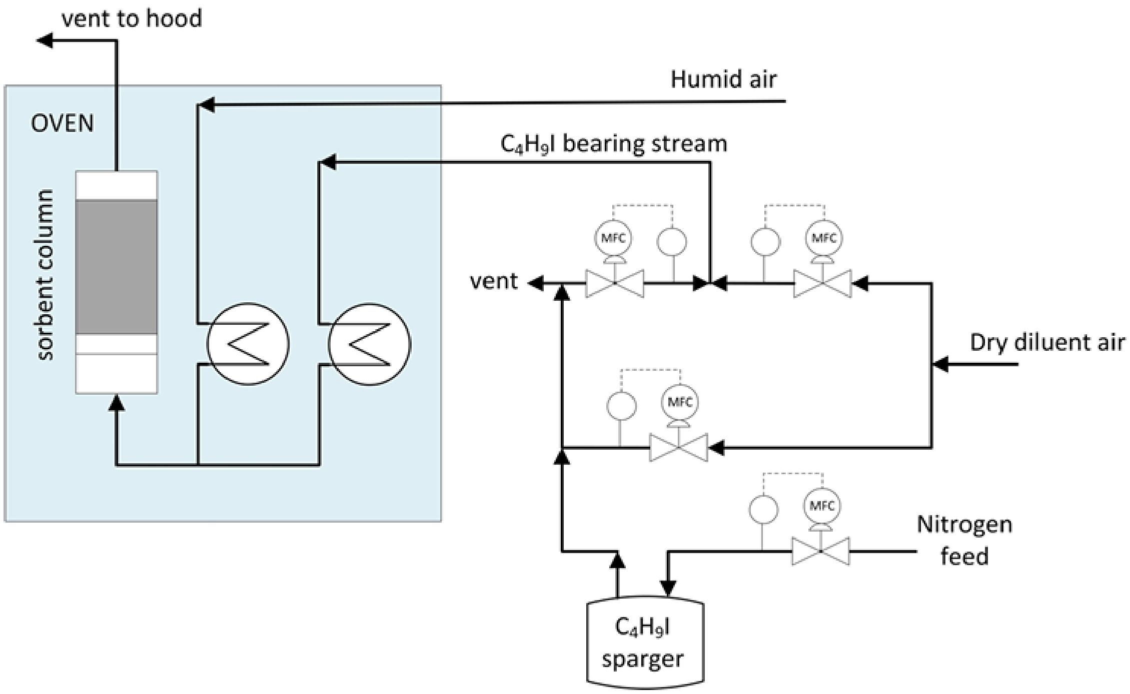


Figure 4-5. Schematic of Oak Ridge National Laboratory deep bed test system (Bruffey et al. 2019).

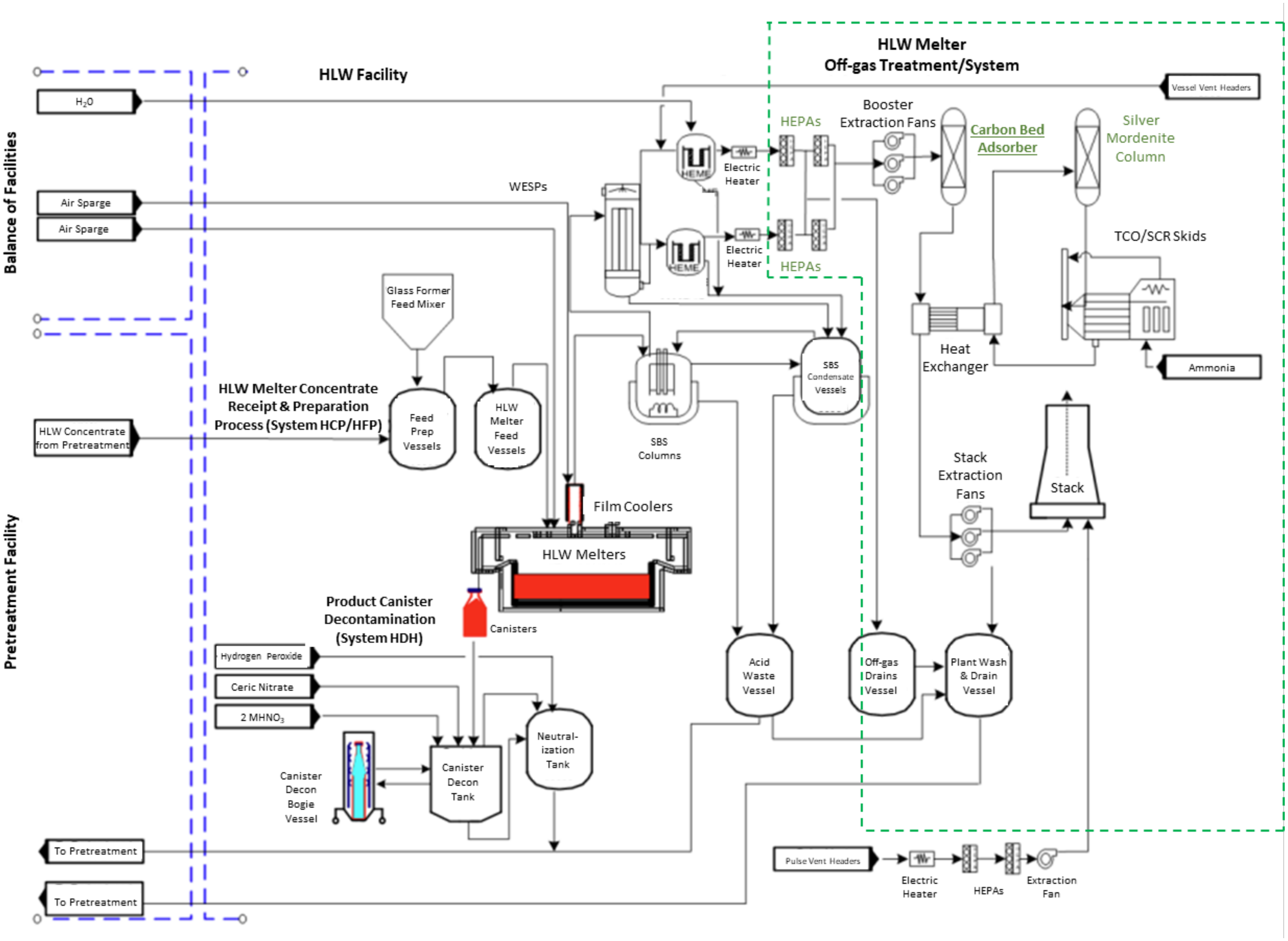


Figure 4-6. Waste immobilization and treatment plant high-level waste vitrification system (WA7890008967).

For chemisorption-based capture sorbents, these technologies work by binding the target analyte and forming a chemically and thermodynamically stable coordination or compound. Examples of this include the reaction of silver with iodine to form silver iodide (i.e., $2\ Ag^0_{[s]} + I_{2[g]} \rightarrow 2\ AgI_{[s]}$) at getter sites within the sorbent. Examples of chemisorption-based sorbents include Ag-mordenite, Ag-faujasite, Ag-aerogel, Ag-xerogel, Clariant AC-6120, Ag-KTB, and Ag-KTC (see Table 4-1). In all these examples, silver is the gettering metal within a matrix form of passive porous substrates. The passive porous substrate acts as a support for holding the active gettering metal (i.e., Ag^+/Ag^0) in place during the capture process. The performances of these sorbents vary under different conditions, including operating temperature, competing gases, and oxidizing conditions. The theoretical capacity potential for a given silver-containing chemisorption-based sorbent is based on the mass of available silver in the sorbent, but that does not mean that a sorbent will operate with full utilization of active getter available. The active getter loading can vary significantly between sorbents (i.e., by an order of magnitude).

The capture efficiency and DF of a particular sorbent will depend on the material characteristics (e.g., morphology, void volume fraction, active site density, and tortuosity), analyte chemistry (competing species, chemisorption vs. physisorption) and operating conditions (e.g., temperature, flow rate, absolute pressure, and relative humidity). The sorbent material may need to be qualified to meet the material characteristics needed to meet a design basis DF per the specific MSR off-gas conditions. To illustrate the effects of two of these factors, commercially available NUSORB® activated carbons were considered, as these have been used for noble gas delay at nuclear power plants. At the same temperature, the dynamic adsorption factor (Dynamic K, a measure of adsorbed volume per mass of sorbent) for Xe is approximately 22 times that measured for krypton over relative humidity ranging from 0 to 60 percent (NUCON 2019). In addition, increased relative humidity is accompanied by a decrease in Dynamic K, as shown in Figure 4-7. Further, decreasing temperature is accompanied by an increase in Dynamic K, as shown in Figure 4-8 (NUCON 2019).

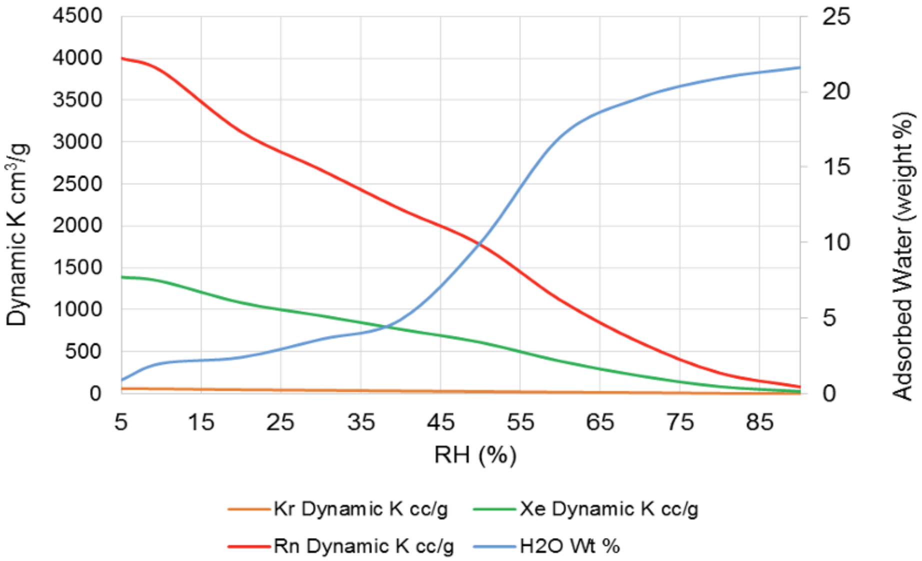


Figure 4-7. Dynamic K for various noble gases versus relative humidity at 20°C (Material: NUCON® NUSORB® Grade GXK™) (NUCON 2019).

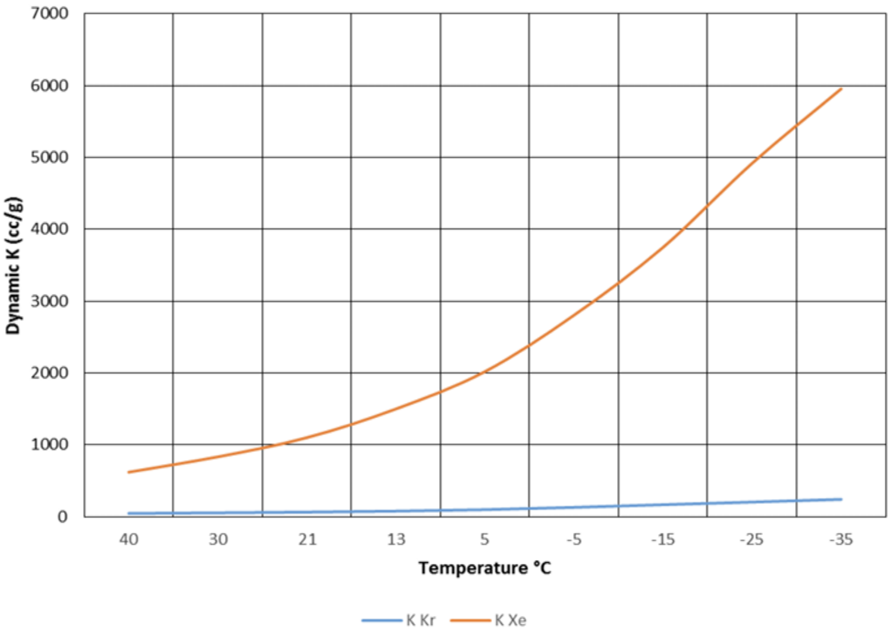


Figure 4-8. Dynamic K for Xe and Kr versus temperature (Material: NUCON® NUSORB® Grade GXK™) (NUCON 2019).

4.2 Safety Considerations

4.2.1 Normal Operations

The most important measurable parameter for the performance of a solid sorbent is the DF across the bed. This can be readily verified through *in situ* (online) monitoring of the pre-bed and post-bed target analyte concentrations (i.e., breakthrough monitoring). Through optical spectroscopies (e.g., ultraviolet, visible, infrared), combined techniques such as laser-induced breakdown spectroscopy, spectrometry methods (e.g., mass spectrometry), and/or radiation detection methods, species concentrations in these streams can be identified and/or quantified *in situ* for continuous monitoring (Andrews et al. 2023; Niu et al. 2022; Robinson et al. 2024; Takeshita and Nakano 2001; Xie et al. 2022). Figure 4-9 and Figure 4-10 illustrate representative spectra generated by some of these techniques for noble gas monitoring and iodine compound monitoring, respectively. Sorbent DFs and lifetimes may be monitored relative to the safety design limits, since (by definition) solid sorbents utilizing chemisorption-based capture processes will eventually become loaded to an extent whereby they can no longer capture radionuclides.

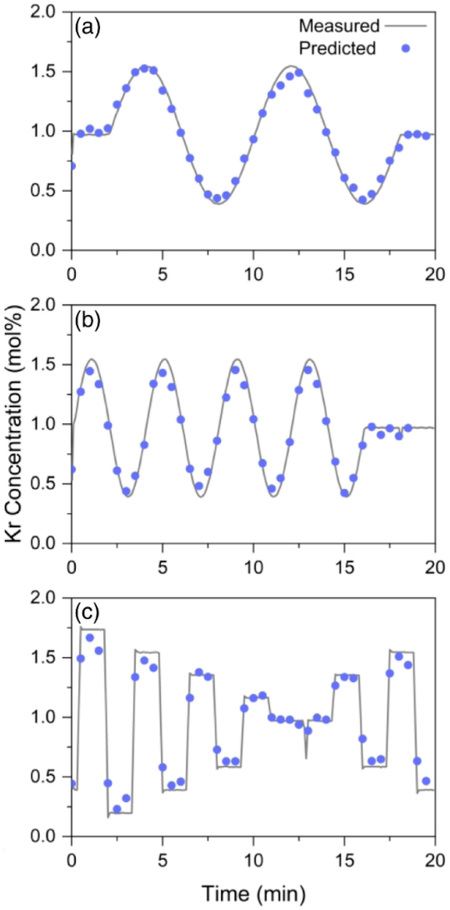


Figure 4-9. Monitoring of noble gas (Kr) concentration by laser-induced breakdown spectroscopy (Andrews et al. 2022).

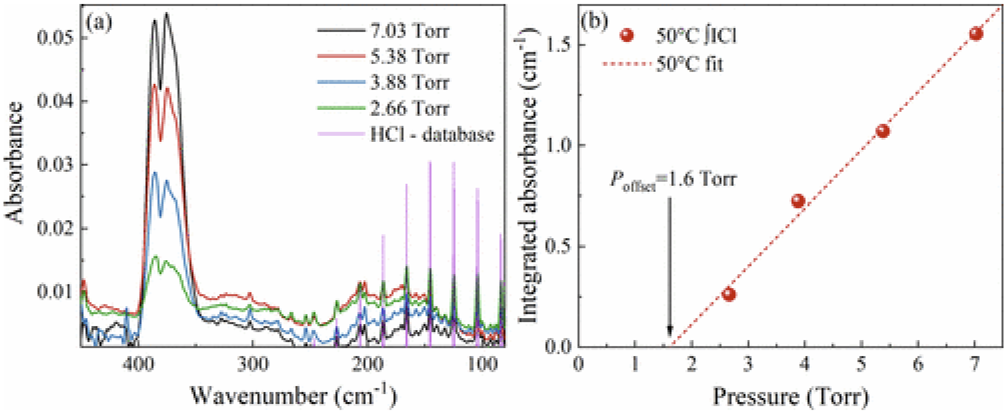


Figure 4-10. (a) Infrared absorbance spectra of iodine monochloride measured at 50°C vs. pressure. (b) Beer's law analysis of the 381 cm^{-1} integrated fundamental vibration band for iodine monochloride (Hughey et al. 2020).

The performance and operability of the solid sorbent bed may be impacted by several issues such as the sorbent becoming saturated or the sorbent being contaminated with unwanted species (e.g., particulates, mists) that would decrease the functionality of the bed or render the bed nonfunctional. Performance monitoring can be performed using *in situ* concentration monitoring of target analytes on the backend of the sorbent bed to verify the desired DFs are met. Reference DFs for some example sorbents are shown in Table 4-3 based on prior experience at spent fuel reprocessing plants, although DF ranges are dependent on the specific design.

Table 4-3. Decontamination factors for example sorbents (Doll et al. 2014).

Solid sorbent type	I_2 Decontamination Factor
Ag mordenite	$\leq 10^4$
Ag-exchanged Molecular sieve	10^3–10^4
Silver nitrate impregnated amorphous silicic acid (AgA)	250
AC-6120 silver nitrate impregnated amorphous silicic acid (Clariant AC-6120)	10^2–10^3
Activated carbon	10–10^3

Other aspects that need to be considered are radiation stability, chemical stability, and mechanical stability. For example, while much is known regarding radiation stability for some of these types of materials (e.g., zeolites) (Riley et al. 2016), little is known about many of the developmental materials such as MOFs. The loading of radioiodine (e.g., ^{129}I, ^{131}I) on iodine sorbents will result in the production of xenon fission products (i.e., ^{129}Xe, ^{131}Xe) through β^- decay; these could diffuse through the iodine sorbent bed and require subsequent capture by the noble gas capture bed. It is unclear how these β^- decay processes will degrade the solid sorbent beds. Zeolites exhibit good chemical stability; exposure to acidic gases (e.g., HCl) may alter their composition by dealumination (hydrolysis of Al–O–Si bonds), which increases Si/Al ratios and increases the specific surface area and mesopore volume (Cakicioglu-Ozkan and Ulku 2005). As noted in Table 4-2, chloride will compete with iodine to form silver chloride for silver-loaded sorbents. The mechanical integrity is also very important, as highly porous supports (e.g., zeolites, aerogels, xerogels) have increased friability due to the increased void volume in their structures. High-carbon sorbents (e.g., activated carbons) are known to become flammable under certain conditions (e.g., highly oxidizing, in fine-dust form), although the minimal concentration of oxidants in the MSR off-gas would not support combustion.

It is anticipated that a molten hydroxide scrubber with incorporated solid-sorbent capture bed system would be in operation whenever the reactor is in operation. In an MSR, off-gases will be continuously produced and, if not sent to an off-gas processing system will collect in the gas headspace (e.g., pump bowl headspace or surge tanks), and may interfere with reactor neutronics (e.g., Xe isotopes), or potentially cause unwanted reactions with components. If the primary system were to be down for some reason (e.g., maintenance or sorbent replacement), a redundant (secondary) system would likely be required to be placed online while the primary system is unavailable. This can be accomplished by a lead-lag configuration, a fully redundant system (e.g., redundant scrubber and sorbent bed system), or both.

Decay heat will need to be managed. This can be minimized by the use of an inert gas (e.g., He, Ar) carrier gas flow, which would maintain temperatures within design limits (Ritzmann et al. 2024; Stephenson et al. 2020). He has a higher thermal conductivity relative to Ar, although the heat transfer coefficient may not be much higher for the turbulent flow conditions in the sorbent bed (Burak and Chen 2024). Temperatures need to be continuously monitored and controlled to avoid solid sorbent material degradation.

The final fates for these solid sorbents will vary, but all introduce potential safety hazards for consideration. Except for MOFs and activated carbons, all the chemisorption-based iodine solid sorbents could be hot pressed into a final form for very long term storage, as further explained below. All these sorbents could be placed inside of a secondary form (e.g., grout, cement) and packaged. For noble gas sorbents (e.g., MOFs, Ag-mordenite, H-mordenite), the concepts being explored assume that the noble gases will remain in interim storage with the sorbents in a compressed gas bottle during the fission product decay process.

4.2.2 Off-Normal Conditions

Plugging of solid sorbent beds would be mitigated by the implementation of upstream prefilter traps (e.g., particulate traps). Loss of power would not affect prefilter traps as they are passive, although power loss would affect electrically-powered components of the sorbent bed (e.g., temperature-controlling or cooling components). Potential off-normal conditions would result in elevated bed temperatures and pressure drops along the bed. Fires are a potential hazard for carbon beds. A hazards analysis for the proposed bed would identify consequences and risks for potential off-normal conditions of the specific design. Table 4-4 provides a partial set of potential off-normal conditions and potential options to address these conditions. These options may or may not require shutdown of the reactor and would consider safety risks for each approach. To monitor the performance of the off-gas processing facility, online monitoring would be performed to track fission products of concern and help prevent unwanted releases. To help mitigate unwanted fission product releases, larger solid sorbent beds could be implemented to increase bed operation lifetimes (i.e., time-to-breakthrough). At lower temperatures, radiolysis and damage to the bed are possibilities.

Table 4-4. Summary of potential off-normal conditions and potential options for solid sorbent based technologies.

Off-Normal Condition	Potential Options
The solid sorbent bed no longer performs as intended due to elevated temperatures, reaching saturation limits, high radiolytic damage to the sorbent bed, plugging of the bed, or other issues leading to lower-than-intended capture bed efficiency. These conditions would result in lower DFs.	The likely action here is a solid sorbent bed replacement with a fresh bed volume (sorbent column replacement). As indicated in Section 4.2.1, the design would likely include a primary and secondary system in series, and the intent would be to switch to the secondary system during sorbent changeout in the primary. It is likely that changeout operations would be conducted remotely (e.g., manipulators).
The solid sorbent bed no longer performs as intended due to rupture of the bed containment (e.g., as a result of corrosion or a contained fire). The condition may lead to release of sorbed species.	In a condition of isolated failure such as that associated with corrosion or a contained fire, the likely action here is a solid sorbent bed replacement with a fresh bed volume (sorbent column replacement). As indicated in Section 4.2.1, the design would likely include a primary and secondary system in series, and the intent would be to switch to the secondary system during sorbent changeout in the primary. It is likely that changeout operations would be conducted remotely (e.g., manipulators).

4.3 Considerations on Waste Products

The selection of a solid sorbent material must be made with the end-of-life state in mind; in other words, the storage and very long term storage pathway may need to be evaluated or considered before a solid sorbent can be implemented. Regarding iodine sorbents, capture media relying on chemisorption are often stable suitable forms for storage. For silver-based solid sorbents, the formed AgI is a stable compound, and the bed would fare well after use. This would also be the case for Cu-based sorbents where CuI is formed, which is a stable compound with a low leaching rate (Riley et al. 2021, Riley et al. 2024). However, for physisorption-based capture media, the adequacy for storage would consider the inevitable desorption that the bed will undergo because molecules are adhered to a surface by relatively weak intermolecular forces, which will depend on storage temperatures.

Regarding solid sorbents for noble gas capture, interim storage after capture is the likely path forward because waste forms for noble gases (e.g., hot-pressed zeolites or impregnated copper) tend to be cost prohibitive and would likely not be needed (IAEA 1980). For the MSRE, charcoal sorbent beds were used to allow decay of xenon radioisotopes for approximately 90 days and krypton radioisotopes for approximately 7 days (Haubenreich and Engel 1970). Thus, only stable or long-lived gaseous nuclides were present in the helium carrier gas at discharge through the stack. The length of time for interim storage may vary for MSRs under development. The decay process during storage will generate stable or long-lived solid products (e.g., ^{133}Cs, ^{135}Cs, ^{85}Rb), which would be retained in the bed. There is some evidence that rubidium may enhance corrosion of steel in nuclear material storage (Asmussen and Neeway 2020); therefore, the potential to degrade the bed containment during periodic use may need to be assessed. Finally, radiolysis of salts and production of corrosive gases (e.g., F_2 or Cl_2) need to be considered for storage of any solid sorbents that may be contaminated with salt aerosols. Online monitoring may be implemented to verify performance of bed containment.

The approach for very long term storage of solid sorbent beds will differ depending on the type of material and the fission product contents associated with the sorbent—see Table 4-5. For zeolite materials, the most likely fate is hot pressing for consolidation and very long term storage. Hot pressing (e.g., hot isostatic pressing or HIP) of chemisorption-based sorbents (e.g., metal-loaded zeolites, aerogels, xerogels, silica, alumina) will result in a passive and durable matrix (e.g., a silica-rich or alumina-rich matrix) surrounding a chemically durable metal iodide (e.g., AgI), sometimes involving collapsing of the pore structure. The point of adding the “hot” part is that this approach will reduce the volume of the sorbent material since the porous nature of the sorbents results in low overall density. The volume reduction will help lower the overall volume of waste requiring very long term storage and would likely reduce costs. The concept of collapsing the pores of the sorbent using hot pressing is also to lower the available surface area for chemical degradation in a condition where the material is exposed to ground water within a very long term storage environment.

Table 4-5. Potential storage and very long term storage approaches for example solid sorbents.

Solid sorbent type	Example sorbent material	Storage and very long term storage approach
Zeolite	Ag-mordenite, Ag-faujasite, H-mordenite	Hot pressing
Molecular sieve	Metal-organic framework (MOF, e.g., HKUST-1, SBMOF-1); zeolites (e.g., silver mordenite, hydrogen mordenite)	Packaging in compressed gas containers or cartridges; Conversion to glass composites
Aerogel or xerogel	Ag-functionalized silica aerogel; Ag-aluminosilicate aerogels/xerogels, chalcogels (sulfide aerogels)	Hot pressing

Solid sorbent type	Example sorbent material	Storage and very long term storage approach
Porous silica	Ag-KTB and Ag-KTC (Ag-loaded silicic acid)(***commercial***)	Hot pressing
Porous alumina	Clariant AC-6120 (***commercial***)	Hot pressing
Carbon	Activated carbon (e.g., BAT-37, Nucon® GXK, DABCO, potassium Iodide [KI], activated carbon)(***commercial***)	Grouting
Desiccant	$CaSO_4$ (***commercial***), MS3A (***commercial***)	Cement and packaging in high-integrity container

Hot pressing using HIP or spark plasma sintering have been demonstrated as consolidation methods for making waste forms for iodine-loaded silver mordenite (Bruffey and Jubin 2015; Riley et al. 2023), silver faujasite (Riley et al. 2023), silver aerogel (Matyáš and Engler 2013; Matyáš et al. 2016), and silver xerogel (Riley et al. 2023). Spark plasma sintering is a laboratory-scale tool, but hot pressing might be used at a larger scale. During hot pressing, it is possible for new structural species to form such as iodine-containing zeolites converting to the sodalite crystalline structure. Iodine-loaded silver-based zeolites contain AgI crystals within and around the zeolite structure, but sodalite is a more robust crystalline structure for preventing the release of iodine as the AgI crystals are encapsulated within a sodalite β-cage (Bruffey et al. 2016; Chong et al. 2020). In addition to just crystal-to-crystal conversions (e.g., zeolite → sodalite) for metal-iodide stabilization, additional options exist for waste form production that include encapsulating the metal-iodide in a secondary matrix such as grout or cement (Kaplan et al. 2019; Li et al. 2019; Fujii et al. 2022), a metal (e.g., a ceramic-metal or "cermet" composite)(Evarts et al. 2024), or a glass (e.g., a glass-bonded ceramic)(Riley et al. 2017a).

For materials like activated carbon, the most likely fate would be a low-temperature consolidation process such as grouting into a monolithic form. For noble-gas capture media, these could be loaded into compressed gas cylinders and left for fission product decay. MOFs could be converted to durable glass-composite forms, e.g., by coating with Bi_2O_3-ZnO-B_2O_3 and Bi_2O_3-ZnO-SiO_2 glass powders (Pénélope et al. 2022). The MOFs, glass powders, and metallic silver may be crushed together and heat treated at 500°C to form dense pellet monoliths. The metallic silver would capture iodine released during the treatment process as AgI. For tritium-containing sorbents, the likely pathway for very long term storage of tritiated water is that these will be captured in desiccants (e.g., $CaSO_4$, molecular sieve 3A or MS3A), cemented, and then stored in high-integrity containers (Vienna et al. 2015).

In the late 1970s at the Idaho Chemical Processing Plant, off-gas from fuel dissolution was treated using cryogenic distillation to remove ^{85}Kr, which was then used in encapsulation studies. The encapsulation test process used solid zeolite 5A material and included hot isostatic pressing. In 2010, five metal capsules were recovered, and in 2014, the preserved capsules were shipped to ORNL for analysis. Of the five capsules, two had been previously breached and were analyzed prior to opening the three intact capsules. One of these two capsules had undergone hot isostatic pressing and the other did not. Results of the two breached capsules showed that there was a relatively uniform distribution of krypton and rubidium within both capsules. There was extensive corrosion throughout the capsule walls and analysis showed the presence of iron oxide. Lead was detected within the capsules that underwent HIP encapsulation, although the origin of lead was unknown (Jubin and Bruffey 2016). X-ray diffraction showed a shift in the 5A structure, which was assumed to be due to the encapsulation/sintering process. Results of this study indicated that even though there was a breach to the capsule wall, a “significant fraction” of the initially encapsulated krypton material remained, which could indicate that both HIP and non-HIP encapsulation methods are promising as a long-term waste form (Jubin and Bruffey 2019).

4.4 Technology Status

4.4.1 Prior Experience

Solid sorbents have been implemented in nuclear processes for many decades. The commonly implemented solid sorbents for nuclear applications include silver zeolites (e.g., Ag-mordenite, Ag-faujasite), silver-loaded alumina (i.e., Clariant AC-6120), silver-loaded silicic acid (e.g., Ag-KTB, Ag-KTC), and activated carbon (Haefner and Tranter 2007; Riley et al. 2016). Additional sorbents less studied include silver-functionalized silica aerogel (Ag-aero)(Matyáš et al. 2018), silver-functionalized aluminosilicate aerogels/xerogels (Riley et al. 2017b, 2022), and chalcogels (Riley et al. 2014; Subrahmanyam et al. 2015).

According to McFarlane et al. (2019c), the MSRE used (and the conceptual MSBR was planned to use) charcoal beds for capture of off-gas components with a decay holdup for 90 days whereby ^{85}Kr was left as the most important source term contributor. The charcoal beds consisted of pipes filled with charcoal, submerged in a water-filled pit at room temperature (Haubenreich and Engel 1970). This system was designed such that a slip stream (10 percent of total salt volume) was bubbled with He to strip off contaminants and fission product gases. Once cleaned, the helium gas supply was returned to the purge system. The salt spray was not well executed and was prone to clogging. Since this time period, global solid sorbent design and testing efforts have been performed to find and develop more effective capture processes for these target radionuclides.

For tritium capture in the form of water, desiccants have been commercially available for many decades and are a mature technology.

4.4.2 Information Gaps or Needs

While hundreds (if not thousands) of solid sorbents have been documented in the literature for radionuclide capture (some including immobilization concepts), many have only been tested under rudimentary conditions that do not reflect actual operating conditions to be expected in a plant and have limited implementation maturity (see Table 4-1). This means that testing protocols usually are performed under static conditions (not dynamic), do not include stream contaminants (e.g., acids and aerosols), and do not include species that can compete for binding sites (e.g., $Cl_{2(g)}$, $Br_{2(g)}$), are not performed under prototypical flow rates and residence times for long time periods or under radiation conditions. Limitations to deployment include cost, scalability (many are only demonstrated at production on the gram scale), and unknown properties (e.g., aforementioned gaps, radiation stability, chemical stability, mechanical stability).

Information is needed on the long-term chemical durability of new materials. Most iodine sorbents deployed today include silver, a metal that is regulated under the Resource Conservation and Recovery Act under 40 CFR Part 261 when disposed. The concentration of silver in final waste forms is unknown, and consequently at this time it is unclear how the silver could affect potential waste management scenarios. Alternative capture materials are being studied across the world to replace silver-based sorbents with some of the more promising materials being Bi-based and Cu-based sorbents (Tesfay Reda et al. 2021; Riley et al. 2021).

While several iodine sorbents are available commercially, the most widely used sorbent for iodine capture in nuclear environments (i.e., silver mordenite) is no longer commercially available. Potentially suitable chemisorption-based sorbent alternatives to Ag-mordenite have been evaluated in the distant past and in recent years that include Clariant AC-6120 and other Ag-zeolites (e.g., Ag-faujasite). While physisorption-based capture materials might be less expensive than silver-based chemisorption-based sorbents, captured iodine would not remain bound to the sorbent for a long period of time simply due to low energy binding mechanism.

Other information gaps to be addressed include relevant testing conditions for candidate solid sorbent materials; this would likely include the implementation of the pre-filter components described upstream of the solid sorbent beds in Figure 1-1. While commercially available and implementation ready technologies do exist for iodine (see Table 4-1), solid sorbents for noble gas capture are not well developed. It is unclear how any of these materials will behave under the MSR off-gas stream, which will be comprised of different constituents than is typically found in reprocessing facility off-gas. It is well known that some silver-containing sorbents can undergo aging effects in prolonged exposure tests under oxidizing conditions, thereby shortening operating life (e.g., Ag-mordenite, Ag-aerogel)(Patton et al. 2014; Nan et al. 2017; Bruffey et al. 2015) but that is likely not a concern for MSRs under normal operating conditions. Running aging studies for candidate solid sorbents would be done under conditions representative of expected MSR operations where commercially available sorbents could be tested first followed by other exploratory materials.

5.0 Delay Systems

5.1 Technology Description

The design principle for a delay off-gas system involves capture and holdup of short-lived noble fission gases using a combination of multi-chamber pathways (e.g., 10-min delay line), delay tanks, and/or solid sorbent delay beds (McFarlane et al. 2020). Normal operation using a delay system involves hold up of various isotopes for timeframes on the order of days (~10 half-lives—see Table 5-1), which would decay and be captured as stable products. For example, activated carbon beds could be used to delay the release of xenon from several weeks to several months (Doll et al. 2014). The remaining longer-lived isotopes would pass through the delay beds and be captured for long-term storage. Additional discussion regarding sorbent materials is found in Section 4.0.

Table 5-1. Summary of short-lived fission products targeted for a decay tank, including the half-life ($t_{1/2}$) of the target radioisotope and the decay products.

Fission Product	Half-life ($t_{1/2}$)	Decay Product(s)
^{139}Xe	39.68 s	^{139}Cs (9.3 min) → ^{139}Ba (1.396 h) → ^{139}La
^{137}Xe	3.82 min	^{137}Cs (30.17 y) → ^{137}Ba (stable)
^{135m}Xe	15.36 min	^{135}Cs (2.3×10^6 y) → ^{135}Ba (stable)
^{135}Xe	9.1 h	^{135}Cs (2.3×10^6 y) → ^{135}Ba (stable)
^{133m}Xe	2.19 d	^{133}Cs (stable)
^{133}Xe	5.243 d	^{133}Cs (stable)
^{131m}Xe	11.8 d	^{131}Xe (stable)
^{89}Kr	3.15 min	^{89}Rb (15.44 min) → ^{89}Sr (50.52 d) → ^{89}Y (stable)
^{88}Kr	2.84 h	^{88}Rb (17.7 min) → ^{88}Sr (stable)
^{87}Kr	1.27 h	^{87}Rb (4.8×10^{10} y) → ^{87}Sr (stable)

Figure 5-1 illustrates an example approach of a delay off-gas system for a medical isotope production facility, the Australian Nuclear Medicine (ANM) Active Ventilation System, which consists of multiple delay tanks followed by carbon-sorbent delay beds. The cover gas is held in the delay tanks for approximately 7 weeks, transferred to carbon beds, followed by release after approximately 15 days. A view of the ANM delay tanks is shown in Figure 5-2 (Hoffman and Brink 2021).

A preferred solid sorbent medium for a delay bed would include the ability for dynamic adsorption, which is the ability to adhere gas molecules to the sorbent, allow for desorption, and would generally have varying adsorption and desorption rates for each gas species depending on their thermophysical properties. Activated carbon (charcoal) has shown to exhibit dynamic adsorption of krypton and xenon and has been used as a medium for delay beds. The holdup time of a charcoal bed is a function of the mass of charcoal, the dynamic adsorption coefficient for each noble gas (Xe, Kr), and the gas flow rate. Testing has shown that the dynamic adsorption coefficient for xenon under operational conditions is approximately 20 times higher than for krypton, thus the xenon holdup time is 20 times longer. Testing has also shown that removal of moisture increases the charcoal adsorption efficiency, which can be accomplished by cooler condensers and desiccant dryers upstream. Figure 5-3 illustrates the use of a delay line and parallel charcoal delay beds in a typical BWR off-gas system (NRC 2012).

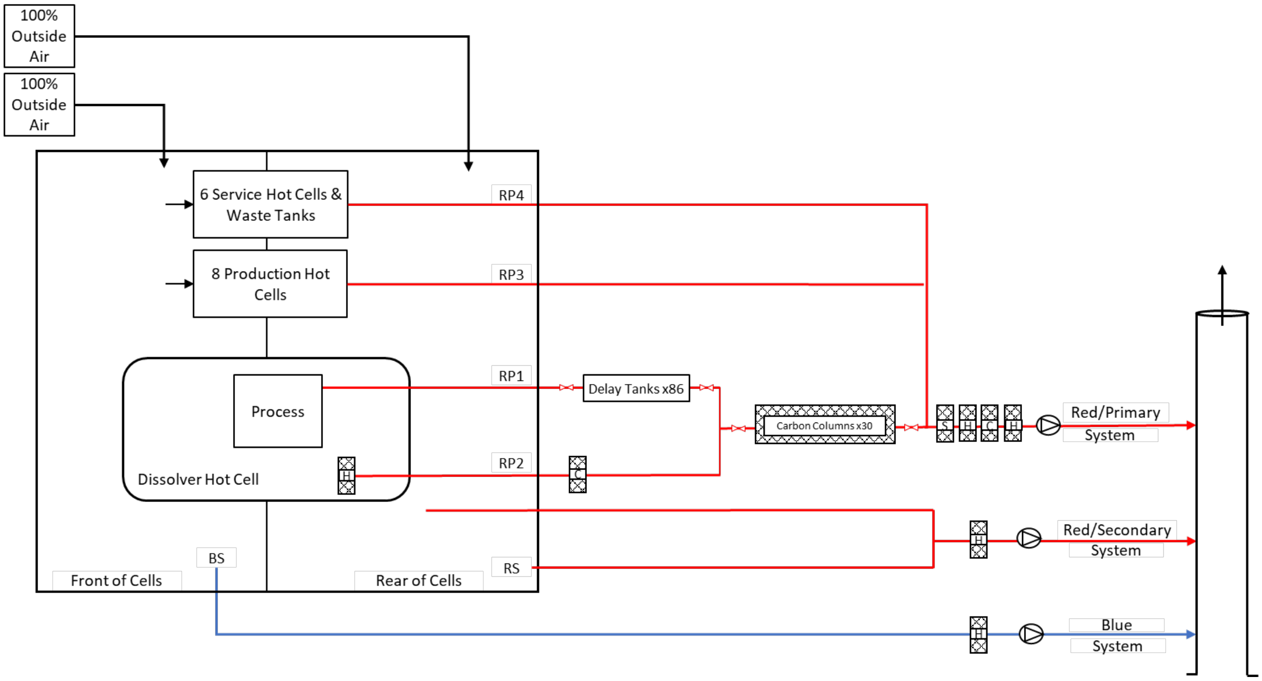


Figure 5-1. Schematic of Australian Nuclear Medicine Active Ventilation System (Hoffman and Brink 2021).

Figure 5-2. Example Delay Tanks from the Australian Nuclear Medicine Active Ventilation System (Hoffman and Brink 2021).

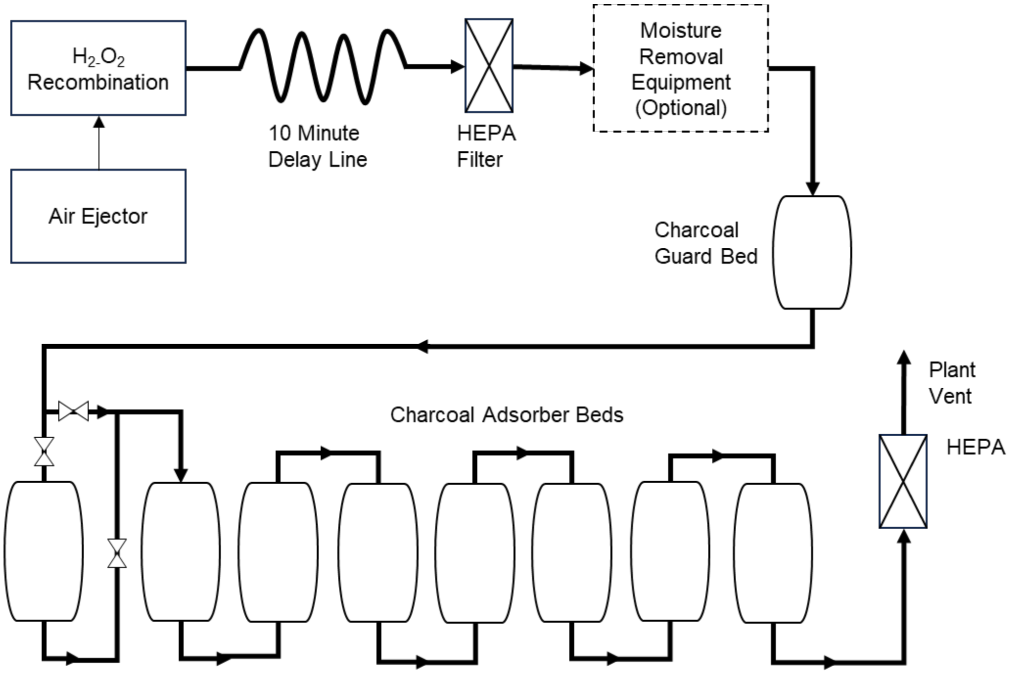


Figure 5-3. Example flowsheet of a delay system using delay lines and charcoal beds in a boiling water reactor off-gas system (reproduced from NRC 2012).

Components and materials for a delay system are commercially available and have been used at medical isotope production facilities (Stephenson 2020), where uranium irradiation and ^{99}Mo extraction include radioxenon as a byproduct. Multiple, large decay tanks and delay beds are often used to reduce emission levels of the off-gas. Figure 5-4 shows a schematic of a general medical isotope abatement process. During this process, off-gas is held in a buffer tank to allow short-lived isotopes to decay and then processed through the decay tanks and delay beds. The Korea Atomic Energy Research Institute has also proposed a design replacing the large decay tanks with cooled adsorbent beds, which allows for longer holdup within a smaller area (Stephenson 2020).

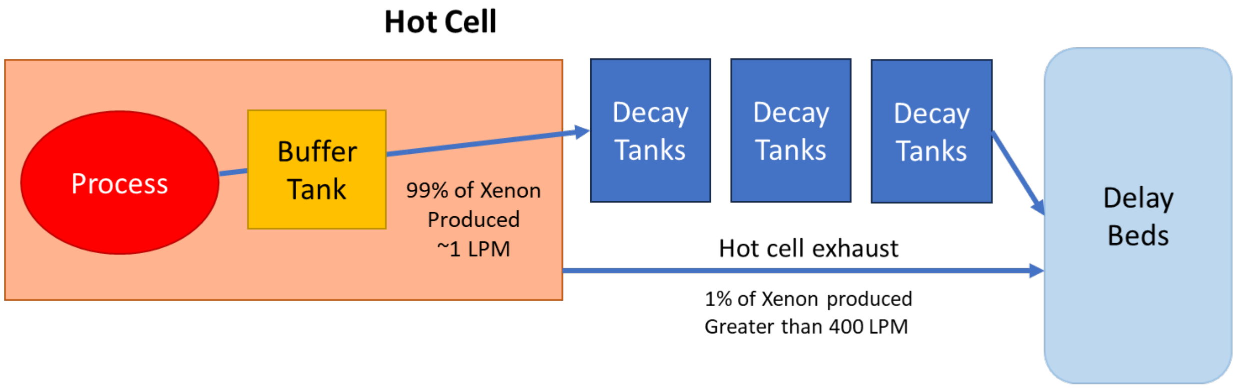


Figure 5-4. Example medical isotope abatement process using decay tanks and beds for xenon decay (Stephenson 2020).

Abatement facilities incorporate very large process tanks loaded with solid sorbents. Increased DFs would require increased charcoal or different sorbent material (e.g., silver zeolite, MOFs), optimized gas flow rates, or potentially cooling of the sorbent material. Typical flow rates for nuclear facilities are in the range of 10^2 to 10^3 L/min, although an MSR facility may have lower flow rates. The MSBR planned to use a helium carrier stream flowing at a rate of 312 L/m (McFarlane et al. 2020). Higher DFs are achieved with lower flow rates through sorbent beds. If flow rates are low enough, it may be possible to use alternate sorbent materials at lower masses and volumes. Systems are generally operated below atmospheric pressure to prevent outward gas leaks. A back-end blower is typically used to route decay gases to the next component of the off-gas treatment system or to the facility vent stack.

5.2 Safety Considerations

5.2.1 Normal Operations

In off-gas management systems that incorporate multiple delay tanks, valves are used to divert gas flow from one delay tank to another as each tank is filled. Where there are multiple tanks, one approach is to monitor the pressure of a tank and when a pressure threshold is reached, indicating the tank is full, automatically switch inlet flow to the next tank. The full tanks are stored until noble gases decay to acceptable activity limits; this storage time could be based on monitoring by gamma spectroscopy or by a verified and validated modeling approach rather than monitoring the tank. Following storage, the contents are routed to the next component of the off-gas treatment system or to the facility vent stack. Continued pressure monitoring of full tanks can also be used to monitor for potential leakage, which could indicate the need for valve replacement or other maintenance activities. If cooling is incorporated (Lee 2021), temperature monitoring of the delay tank contents would be required in addition to pressure monitoring.

Performance monitoring may be implemented to ensure that the sorbent bed is functioning properly, e.g., noble gases are not getting through the bed prematurely or potential degradation phenomena (e.g., erosion, radiolytic damage of MOFs). It is presumed that off-gas monitoring could be performed by monitoring pressure drops across the bed or by using a qualified in-situ detection process such as a mass spectrometer or real-time γ-measurements of the exiting stream. These techniques may not be able to identify the root cause of the problem but serve to identify a need to divert flow to a secondary bed.

Potential lifecycle and maintenance concerns for the delay tanks include fouling from non-volatile decay products and long-term exposure to considerable dose. While the dose from radioxenon and radiokrypton in the delay tanks is quite large, the actual mass passing is quite small. For instance, processing 10,000 Ci/day (3.7×10^{14} Bq/day) of ^{133}Xe amounts to 53 mg/day or 20 g/yr. For large abatement tanks (e.g., a tank loaded with 10 m^3 or 5 metric tons of activated charcoal), this is a very small fraction of the sorbent mass. For comparison, the dynamic adsorption coefficient for xenon on commercially-available NUCON® GXK activated carbon (NUCON 2019) is 767 cm^3/g or 4.6 g/g (grams ^{133}Xe per gram of sorbent). Therefore, based on the mass of material that could plate to the sorption capacity of the sorbent material, it may be concluded that plating of non-volatile decay products (i.e., Cs, Ba, Rb, Sr, Y) will have a minimal effect on the DF for the system. The amount of plating from decay products will scale with the amount of krypton and xenon produced, but a relatively large increase would be required to create an issue over the lifetime of the bed.

5.2.2 Off-Normal Conditions

Off-normal conditions may lead to increases in emissions of various radioisotopes. For delay tanks, the major concern involves the temperature rise in the early stages of the abatement bed (or in the first bed of a multi-bed abatement system) for a sufficiently large increase in off-gas emissions from the reactor. Absent redundant containment measures downstream from the delay tanks (e.g., by diverting flow to backup/redundant delay tanks), the radioactive emissions at the end of the delay train could be elevated above their baseline levels for hours-to-days. Modeling suggests that DFs will not change substantially as a result of these conditions; however, there will be a period of elevated emissions (Ritzmann et al. 2024; Stephenson et al. 2020).

Elevated temperature conditions may increase the pressure inside the delay tank, which can be accounted for in the design. However, such conditions have the potential to push activated carbon beds above their ignition temperature and lead to fire if sufficient oxygen is present in the carrier gas. The potential for fire ignition of activated carbon beds could be evaluated if the reactor design is able to demonstrate insufficient oxygen in the carrier gas. Other materials considered for delay tanks may be relatively more stable at elevated temperatures (e.g., zeolites, molecular sieves, MOFs) with a lower risk of ignition, depending on the polymer-based engineered form used. However, these materials would require significantly more development relative to use of activated charcoal, which has been implemented at nuclear facilities. Elevated temperatures may also lead to structural deformation of the polymer-based engineered form and changes to the void volume in the material, which may impact measurable DFs.

Other off-normal conditions could include the unexpected release of noble gases through the bed, reduced bed capture efficiency, or buildup of nongaseous daughter products in the bed. The safety analyses may need to consider the spontaneous complete release of the gases and volatiles in the decay tank system to define a bounding transient activity limit during an accident (e.g., releases curies/becquerels per day). A hazards analysis for the proposed delay system would identify consequences and risks for potential off-normal conditions of the specific design. Table 5-2 provides a partial set of potential off-normal conditions and potential options to address these conditions. Similar to normal operations, gamma spectroscopy could be implemented for detection of off-normal conditions.

Table 5-2. Summary of potential off-normal conditions and potential options for delay beds and tanks.

Off-Normal Condition	Potential Options
Unexpected spike in off-gas stream flow rate (e.g., due to power level changes in reactor) leading to lower DFs.	Divert flow to backup/redundant delay tanks or bed.
Solid sorbent in the delay bed no longer functions as intended due to elevated temperatures, reaching saturation limits, high radiolytic damage to the sorbent bed, plugging of the bed, or other issues leading to lower-than-intended DFs.	Divert flow to backup/redundant delay tanks or beds. Replace solid sorbent bed.
The solid sorbent bed no longer performs as intended due to rupture of the bed containment (e.g., as a result of corrosion or a contained fire). The condition may lead to release of sorbed species.	Divert flow to backup/redundant delay tanks or beds during valve maintenance or replacement.

In a condition where the system is plugged, a backup needs to be ready for insertion in its place. The maintenance required for such a delay system would typically warrant redundancy. Many delay tank designs could incorporate solid sorbents; additional information for off-normal conditions related to solid sorbents is found in Section 4.0.

5.3 Considerations on Waste Products

Waste management approaches can be developed to optimize the waste treatment from multiple streams within the plant as shown in Figure 1-2. Delay beds are specifically designed for short-lived radionuclides and could even be in place for the life of the facility. Residual species in the activated charcoal are largely expected to be stable, and are expected to be below Class C low-level waste limits for disposal. However, relocation of sorbed actinides may need to be assessed to demonstrate criticality safety for disposal. In the case of MSRs, the final waste classification will depend on the loading of radioactive fission products that are trapped in the delay bed.

Replacement of spent charcoal may occur to maintain treatment efficiency (e.g., moisture exposure can result in a deterioration of performance, although expected to be minimal for MSRs). As an operational example, the Hanford Waste Immobilization and Treatment Plant includes sulfur-impregnated carbon bed adsorbers in the Low Activity Waste off-gas systems, which are used to collect, cool, and treat off-gas from the melters (Ecology 2003; Marcial et al. 2024). The carbon beds are arranged in series and are used to remove mercury and halides from the off-gas. Spent carbon will be replaced periodically and is expected to designate as low-level hazardous waste.

In another design example, the Korea Electric Power Corporation prepared a design certification for the NRC for a light-water reactor. The off-gas system for this design includes gas dryers, two charcoal guard beds, four charcoal delay beds and one high-efficiency particulate absorbing (HEPA) filter. For the possible condition of moisture contamination of the charcoal, a nitrogen purge was included to dry the beds. The charcoal is not expected to be replaced during operation, but if charcoal waste is generated, the plant’s solid waste management program will guide disposal (Korea Electric Power Corporation 2018).

Finally, as previously discussed, the decay process during storage will generate stable solid products ^{85}Rb, which would be retained in the delay bed. Rubidium is known for its role in enhancing corrosion of steel in nuclear material storage (Asmussen and Neeway 2020); therefore, the potential to degrade the bed or waste package containment may need to be assessed.

5.4 Technology Status

5.4.1 Prior Experience

In the MSRE, the main off-gas system was designed with water-cooled charcoal beds to allow a 90-day hold time for xenon and an approximately 7-day hold time for krypton. After delay, the effluent passed through filters and was vented to the atmosphere (Haubenreich and Engel 1970; Chisholm et al. 2020). Delay beds have also been implemented in medical isotope production facilities and operating boiling water reactors such as those in Susquehanna Steam Electric Station and Limerick Generating Station in the United States.

5.4.2 Information Gaps or Needs

The technical maturity of charcoal sorbents is well-established; however, there is far less known about the durability in delay bed applications and the effect of co-contaminants. Additional information is needed to determine carbon changeout frequency.

The release profile of constituents in the MSR carrier gas is important. Steady-state operations of delay beds are well advanced. Delay beds were previously implemented in the MSRE; however, the bed materials and operations were not optimized. Understanding of the interactions of the non-volatile decay products within the sorbent of the delay bed is limited. High performance detectors may help identify location of releases and species released.

Large off-gas streams for delay beds in nuclear facilities are operated near room temperature. MSR outlet temperatures are high (650–700°C), although the off-gas would be initially scrubbed at temperature near 200°C; therefore, cooling of the off-gas is expected prior to a delay bed to improve retention. Additional information could confirm that cooling in holding tanks prior to the charcoal bed would increase retention effectiveness.

6.0 Particle Traps (HEPA Filters)

6.1 Technology Description

Particle traps are used within an off-gas treatment system to remove aerosols, salt mists and splashes, particles/particulates, and condensed daughter products of noble gases. In an MSR, these are typically generated by the fuel salt through splashing or volatilizing (McFarlane et al. 2020). It is likely that these types of species will be present in the high-flow gas stream entering the off-gas treatment system. Removal of these particle types will aide in allowing the other off-gas systems to function properly. Failure to remove these types of particles could lead to degeneration, plugging, and/or contamination of the downstream off-gas treatment units. Particle traps may consist of settling zones and organic (synthetic polymer, carbon) or inorganic (ceramic, glass) media filters.

HEPA filters are used as a basic component to separate suspended particles from the air. They consist of a pleated material fabricated in rectangular or cylindrical shapes, with a particle removal efficiency no less than 99.97 percent for 0.3 µm particles. HEPA filter frames may be manufactured from fire retardant wood or mild, stainless or chromized steels (IAEA 1987).

Air filters can be constructed with either woven or non-woven fibrous or porous particulate media. Non-woven fibrous materials are the most prevalent and can include natural plant-based, regenerated cellulose, natural animal-based keratin, synthetic polymer, carbon, ceramic, mineral, glass, and quartz fibers. Non-woven glass fiber filter paper is the most common material used in industrial HEPA filters (Beckman et al. 2022).

Fiber diameter and type affect filtration efficiency and flow resistance. Regenerated and synthetic fibers are available in various sizes whereas natural cellulose and keratin have limited sizes. Figure 6-1 illustrates the visual differences between different diameter fibers and the effects that diameter size has on filtration efficiency and air flow resistance (Beckman et al. 2022). Table 6-1 shows the typical diameters for different types of fibers.

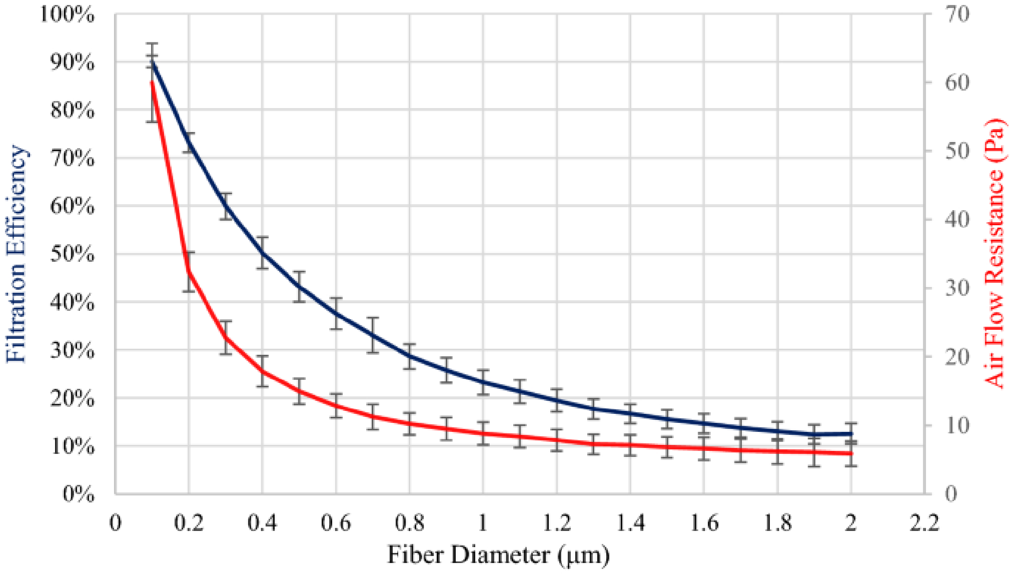


Figure 6-1. Comparison of diameter sizes on filtration efficiency and air flow resistance (Beckman et al. 2022).

Table 6-1. Comparison of example fiber diameter sizes (Beckman 2023).

Fiber	Category	Typical Fiber Diameter (µm)
Carbon Nanotube	Carbon	0.001 to 0.005
Toyobo Zylon PBO	Synthetic Polymer	1.2
Honeywell Spectra 1000 UHMWPE	Synthetic Polymer	1.7 to 3.1
Alumina Saffil	Ceramic	3
E Glass (Alumina Borosilicate)	Glass	3 to 20
Toray T1100G Carbon Fiber (Polyacrylonitrile)	Carbon	5
Fortisan Viscose Rayon	Regenerated Cellulose	9
Quartzel Fibers	Quartz	9 to 14
Basalt Fibers	Basalt	10 to 20
Solvay P25 Carbon Fiber (Pitch)	Carbon	11
American Uppers Cotton Fibers	Natural Cellulose	11 to 22
Dupont Kevlar 49	Synthetic Polymer	12
Oak Wood Fibers	Natural Cellulose	13
Nylon 6 Fibers	Synthetic Polymer	14
Polyacrylonitrile	Synthetic Polymer	16
Wool Fibers	Natural Keratin	18 to 44
Polypropylene	Synthetic Polymer	38
Human Hair for Comparison	Natural Keratin	50 to 70
SCS Ultra^{TM} Silicon Carbide Fiber	Ceramic	75
Boron Fiber	Mineral	142

Tensile strength of the filtration media is specified by American Society of Mechanical Engineers (ASME) to guarantee filters are not easily ripped or torn by accident or during normal operations (e.g., heated, wet, irradiated). Figure 6-2 provides a comparison of tensile strength of select fibers.

HEPA filters trap particulates by forcing the air through the medium where particles either stick or embed themselves directly into the fibers through interception, impaction, and Brownian diffusion (Figure 6-3). As additional particles collect, particle collection shifts to cake filtration where particle deposition accelerates and increased pressure drops, due to particle retention, until the filter reaches terminal design airflow resistance (DOE 2003; Winegardner 1993).

Operational DFs of HEPA filters will be affected by humidity and/or high temperatures. The impact to DF is dependent on the type of media. ASME has mandated, although not an NRC requirement, that filter media be tested to determine resistance to high temperatures and water repellency. For ASME AG-1, filters are required to “withstand a sustained pressure of 2,400 Pa while subjected to continuous heated air flow of 370 °C.” For humidity ASME AG-1 metrics, filters must meet a required average water repellency of at least 5,000 Pa. Figure 6-4 illustrates the temperature limitations of different non-woven fibers. Figure 6-5 provides a comparison of the ability of different non-woven fibers to absorb moisture (Beckman et al. 2022). Moisture regain indicates a fiber’s ability to absorb moisture, expressed as a percentage of water weight absorbed by the fiber compared to the dry weight of the fiber. The influence of moisture on filtration processes will depend on various parameters, including moisture content of the upstream gas, the hygroscopicity of the particles, their size, their chemical composition, and the composition and specific design of the filter. Generally speaking, moisture is expected to reduce capture efficiency of HEPA filters. However, the MSR off-gas would have very low levels of moisture, which would likely not affect HEPA filter performance. Reactor off-gas systems also often use de-misters as a first stage in the filtration process to protect filters from plugging. Table 6-2 shows an overall comparison of fiber operability.

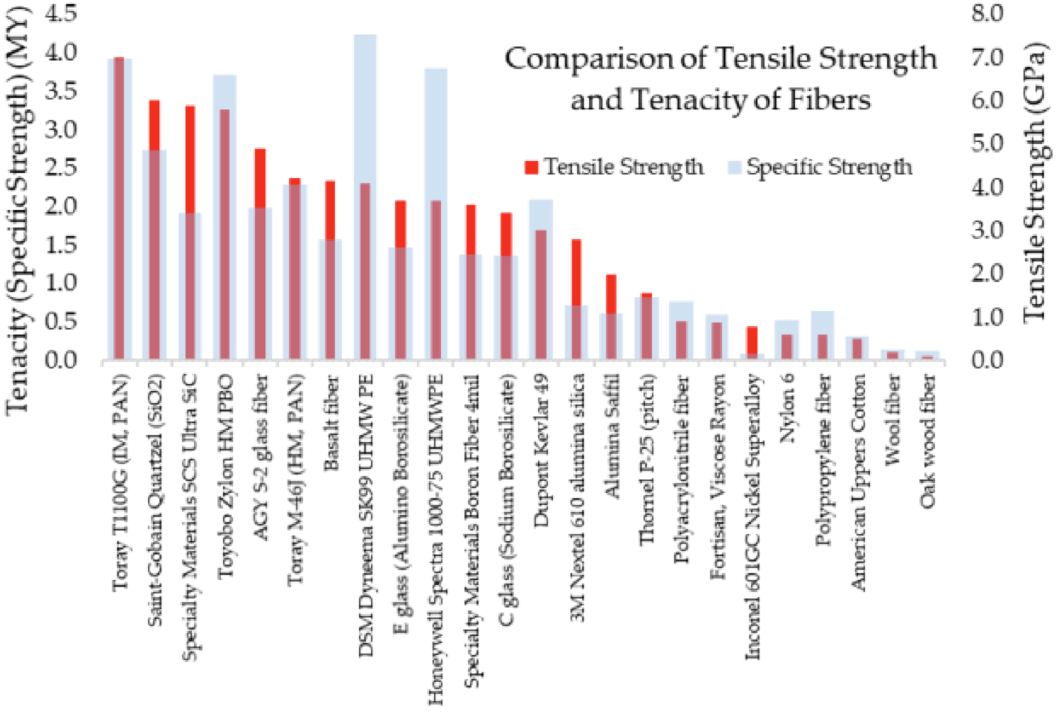


Figure 6-2. Comparison of tensile strength and tenacity of selected non-woven fibers (Beckman et al. 2022).

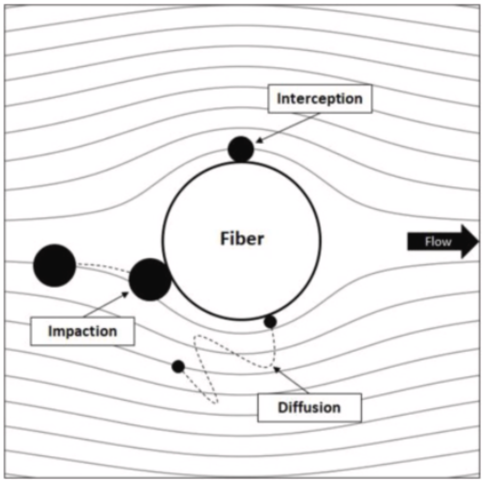


Figure 6-3. Common methods for high-efficiency particulate absorbing (HEPA) filters to trap particulates (Berry et al. 2023).

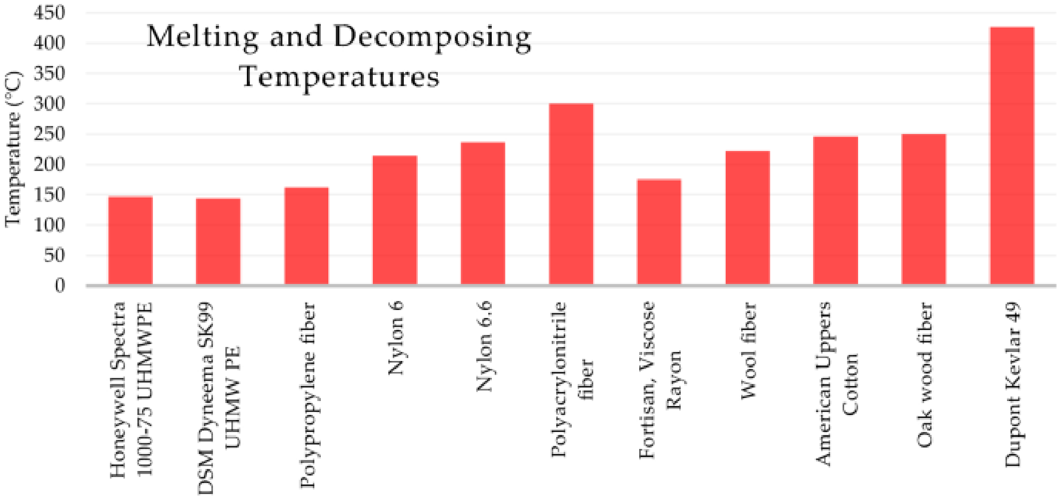


Figure 6-4. Comparison of melting temperatures of select non-woven fibers (Beckman et al. 2022).

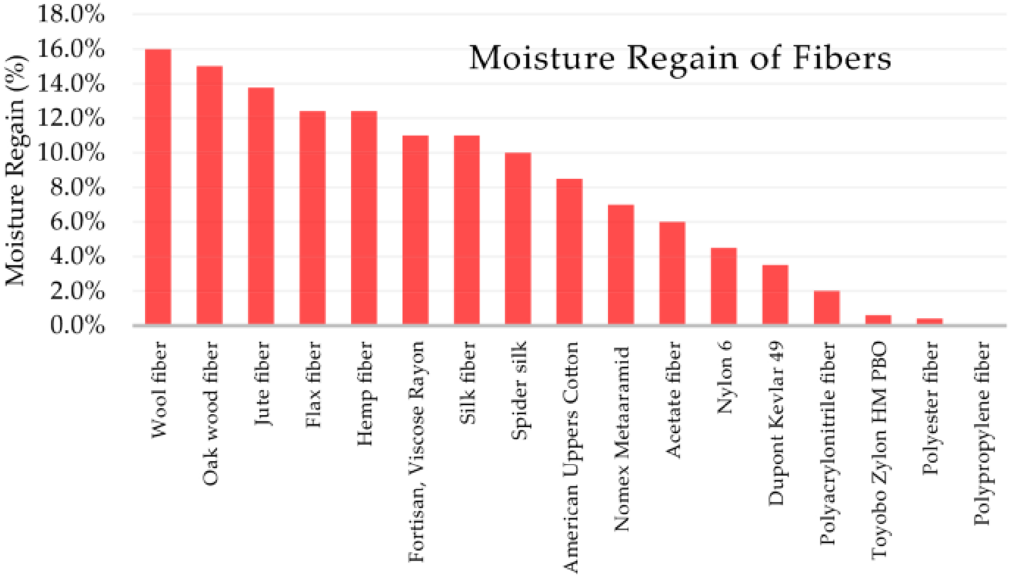


Figure 6-5. Ability of select non-woven fibers to absorb moisture (Beckman et al. 2022).

Table 6-2. Subjective fiber comparison (Beckman 2023).

Fiber	Category	Tensile Strength	Tensile Modulus	Bending	Moisture Regain	Thermal Expansion	Temp 370°C
Cotton	Cellulose	Fair	Elastic	Flexible	Very High	Low	Decompose
Wood	Cellulose	Fair	Elastic	Flexible	Very High	Low	Decompose
Rayon	Regenerated	Fair	Elastic	Flexible	Very High	Low	Decompose
Wool	Keratin	Fair	Elastic	Flexible	Very High	Low	Decompose
Spectra, Dyneema	UHMWPE	High	Stiff	Moderate	None	Negative	Melt
Nylon 6/6.6	Polymer	Fair	Elastic	Flexible	High	Low	Melt
Dupont Kevlar	Aramid	Moderate	Stiff	Moderate	Moderate	Negative	Decompose
Toyobo Zylon PBO	Polymer	Very High	Stiff	Rigid	Moderate	Negative	Near Max
Polyacrylonitrile	Polymer	Fair	Stiff	Moderate	Moderate	Low	Melt
Polypropylene	Polymer	Fair	Elastic	Flexible	None	Low	Melt
Toray M-46J (PAN)	Carbon	High	Most Stiff	Most Rigid	None	Negative	Safe
Toray T1100G (PAN)	Carbon	Very High	Stiff	Rigid	None	Very Low	Safe
Solvay P-25 (pitch)	Carbon	Fair	Stiff	Moderate	None	Very Low	Safe
Saffil	Ceramic	Moderate	Stiff	Rigid	None	Low	Safe
3M Nextel 610 Al Si	Ceramic	Moderate	Very Stiff	Rigid	None	Low	Safe
SCS Silicon Carbide	Ceramic	Very High	Very Stiff	Highly Rigid	None	Low	Safe
Quartzel, Astroquartz	Quartz	Very High	Stiff	Moderate	None	Very Low	Safe
S2 glass fiber	Glass	High	Stiff	Moderate	None	Very Low	Safe
E glass	Glass	High	Stiff	Moderate	None	Low	Safe
C glass	Glass	Moderate	Stiff	Moderate	None	Low	Safe
D glass	Glass	Moderate	Stiff	Moderate	None	Low	Safe
Boron 4 mil	Mineral	Moderate	Very Stiff	Highly Rigid	None	Low	Safe
Basalt	Volcanic	High	Stiff	Moderate	None	Low	Safe
Inconel 601GC Nickel	Metal	Fair	Stiff	Rigid	None	Moderate	Safe

PAN = polyacrylonitrile; PBO = poly (p-phenylene-2, 6-benzbisoxazole); GC = grain-controlled; UHMWPE = ultra-high molecular weight polyethylene.

6.2 Safety Considerations

6.2.1 Normal Operations

For off-gas systems in reactors, the service life of HEPA filters can be monitored by system pressure. Upstream pre-filters, which are often of a type similar to HEPA filters, can be used to prolong the service life of the main filter (IAEA 1987). Filters will have to be swapped out periodically as the capture media becomes plugged and loses operating efficiency. The process is to measure the pressure difference across the filter and, once a threshold is achieved, the gas stream is diverted to a parallel redundant path and the filters are changed. The expected differential pressure operating range will be design-specific.

As described in Section 6.1, optimal performance of filter media is subject to relative humidity and temperature limitations. Consequently, in addition to continuously monitoring pressure drop across the filter unit, the gas upstream would be monitored continuously for temperature and relative humidity; relative humidity would be low for any MSR off-gas. The operating ranges will be design-specific.

Some experimentation has been done to determine the effects of radiation exposure on HEPA filters. An experiment at the Savannah River Site found that γ-irradiation of test-scale HEPA filters only slightly impaired the performance of new filters and temporarily improved the performance of service-aged filters (Jones 1974). A report from the Hanford Site documented that some HEPA filters may contain organic materials of construction that are susceptible to radiation induced damage, including gaskets, wood casing, glue, water repellent, and separator materials (Zavadoski et al. 1996). More recently, DOE documented that the A and B underground filters at Hanford’s B-Plant experienced radiological degradation from excessive radiation exposure (DOE 2003).

Pressure drop across the filter will be an indicator of the performance and percentage of utilized lifetime of the particulate filtration system being implemented. The filters will potentially contain high-dose materials, particularly under off-normal conditions, potentially requiring the ability to change filters without exceeding worker dose thresholds.

6.2.2 Off-Normal Conditions

An off-normal condition could be a sudden loss in pressure drop where a hole might be created within the filtration system. HEPA filters are usually constructed from fiberglass. Table 6-3 provides a partial set of potential off-normal conditions and potential options to address these conditions. A hazards analysis for the proposed particle traps would identify consequences and risks for potential off-normal conditions of the specific design.

Table 6-3. Summary of potential off-normal conditions and potential options for particle traps.

Off-Normal Condition	Potential Options
Inadequate or improper emplacement of filter at installation, leading to a pressure drop, loss of capture efficiency and/or release of filtered particulate.	The filter bed would have to be inspected to identify filter(s) needing re-emplacement or replacement. If the system design consisted of a primary and a secondary (backup) filter train unit, the operator could switch to the other unit while replacement activities were conducted.

Off-Normal Condition	Potential Options
Particulate filters can become plugged over time, leading to a pressure drop and loss of capture efficiency.	The filter bed would have to be replaced to maintain the targeted capture efficiencies. If the system design consisted of a primary and a secondary (backup) filter train unit, the operator could switch to the other unit while replacement activities were conducted.
A rip or tear within the filter media leading to loss of capture efficiently and higher particulate fractions in the downstream gas.	Multiple filters are operated in series and in parallel.

Plugging or damage to the filter are possibilities. If these filters are used before the off-gas sorbent columns, it will be the upstream component to the other off-gas system filtration systems. If a molten hydroxide scrubber (liquid-type) pre-scrubbing bed is not implemented in the off-gas capture (separations) system, then damage to the particle trap would mean potential (and likely) damage to downstream components if this system fails.

The primary gas movers (blowers) will be used to maintain flow through the filters. Filtration systems for the primary reactor off-gas stream would be switched to backup power in a condition of power loss.

6.3 Considerations on Waste Products

Following removal from service, particle traps will be packaged for transportation and possibly stored onsite for a short time period prior to shipment offsite for treatment and disposal as low level waste. Particle traps typically have too much void volume for direct disposal, and consequently require treatment prior to disposal at a licensed commercial facility. This will likely be realized through compaction and possibly grouting the compacts into a final waste form. There is the possibility of waste management optimization that may include filter leaching to reduce the overall filter classification for lower cost handling and disposal. Figure 1-2 shows a flow diagram of how waste streams can be grouped together for disposal.

6.4 Technology Status

6.4.1 Prior Experience

HEPA filters are typically used here to achieve desired particle removal (~99.95 percent) of certain sizes (e.g., 0.3 μm) and are recommended by IAEA for use in nuclear power plants (IAEA 1987). HEPA filter technology has commonly been used in the nuclear industry, including LWRs. NRC’s Regulatory Guide 1.140 Revision 3, “Design, Inspection, and Testing Criteria for Air Filtration and Adsorption Units Of Normal Atmosphere Cleanup Systems In Light-Water-Cooled Nuclear Power Plants,” (NRC 2016) provides guidance on atmospheric cleanup systems in LWRs. Regulatory Guide 1.140 states that light-water reactors would be designed in accordance with ASME AG-1b-2009, “Code on Nuclear Air and Gas Treatment.” Components are recommended to include HEPA filters and prefilters, in addition to activated carbon adsorbers as needed for iodine removal, and heating or cooling elements to control humidity (NRC 2016). HEPA filters were also included in the design for the ORNL MSRE, which was operated from 1965 to 1969 to demonstrate source term calculations (Figure 6-6)(Beall 1964). The DOE has also developed standards and recommendations for use of HEPA filters in nuclear facilities (DOE 2003, DOE 2015).

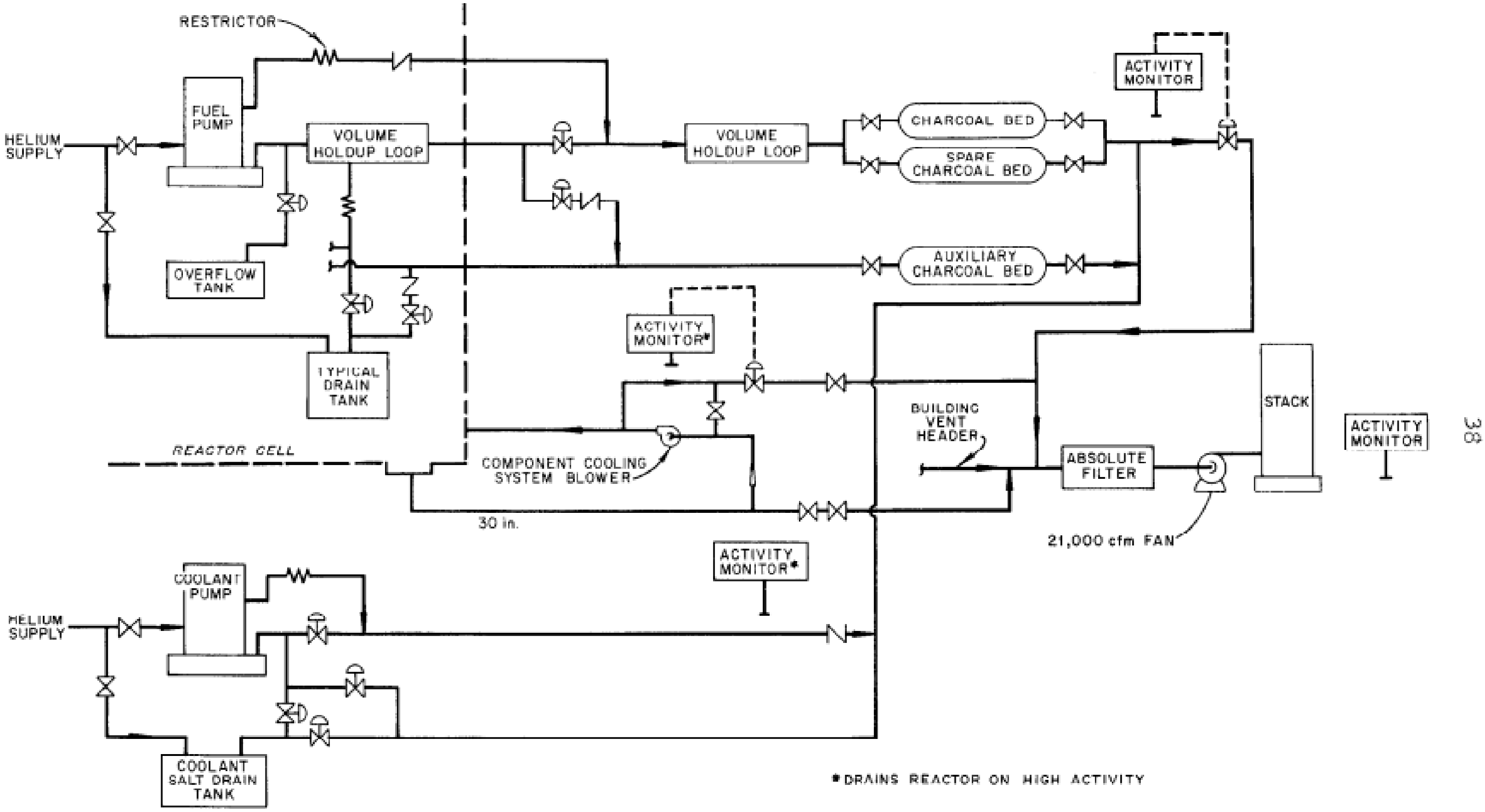


Figure 6-6. Molten salt reactor experiment off-gas system flow diagram (Beall 1964).

Filtration systems are also used in medical, industrial, and academic applications. The IAEA developed Specific Safety Guide No. SSG-59, “Radiation Safety of Accelerator Based Radioisotope Production Facilities,” to provide recommendations for facilities that produce and process radioisotopes. In this safety guide, the IAEA recommend that the production line would be in a clean room or isolator. Filtration and trapping systems would be in place for confining radioactive and hazardous releases from the room before release into the environment (IAEA 2020).

6.4.2 Information Gaps or Needs

Implementation maturity for particulate traps is considered high, as evidenced by wide-scale deployment across the world for a range of applications, including nuclear. Although HEPA filters are common, research continues on damage from moisture, fire, chemicals, and high pressure. Lawrence Livermore National Laboratory (LLNL) has developed the Ceramic HEPA Filter Program to design a nonflammable, corrosion resistant, ceramic HEPA filter in an effort to increase life span and reduce life-cycle cost. In 2019, LLNL fabricated a prototype that includes cylindrical tube ceramic filter elements (Figure 6-7). Preliminary results show ceramic filter elements that have structurally survived tests with 500°C exposure temperatures and demonstrated the capability to exceed performance of many filters related to pressure drops (Haslam 2019). LLNL has also developed ceramic fiber nonwoven media using mini-tubular construction, which shows the potential for use in HEPA filters (Beckman 2023).

HEPA filter testing is also conducted at the Mississippi State Institute for Clean Energy Technology (ICET). At the ICET facility, the Radial Flow Large-Scale HEPA Filter Test Stand (RLSTS) and the Axial Flow Large-Scale HEPA Filter Test Stand (ALSTS) were developed to evaluate nuclear-grade HEPA filters. The RLSTS and ALSTS evaluate performance of HEPA filters on a variety of aerosols with temperatures up to 82°C (77°C for the ALSTS) and humidity up to 90 percent (Mississippi State University 2017). In 2017, the ICET reported on initial evaluations performed on aged filters using the ALSTS, assessing for potential trends in filter failure, tensile strength and moisture repellency. Additional tests are anticipated to be completed to establish a statistically significant amount of data (Waggoner 2017). Additional research on filter material has been recommended by the U.S. Army Corps of Engineers to develop alternate media for HEPA filters (Table 6-4).

Figure 6-7. Lawrence Livermore National Laboratory ceramic high-efficiency particulate absorbing (HEPA) filter design and prototype showing the as-fabricated Class 9 ceramic HEPA filter prototype including (left) design model, (middle) top of filter elements, and (right) side view of the housing prior to assembly (Haslam 2019).

Table 6-4. Recommended material research in filter media (Beckman 2023).

Fiber Type	Further Research Recommendation
Glass	Testing additional glass fiber types (A, C, E, ECR, S2, R, etc.) Production methods to reduce fiber diameter size
Basalt	Production of nonwoven continuous basalt fibers in filter media
Carbon	Electrospinning carbon precursors for air filtration media Carbon nanotube filtration in free molecular flow regime
Ceramics	Electrospinning of sol-gel nonwoven ceramic filter media
Metals	Advancing sintered metal filter media

Radiation damage is an important consideration to replacement schedule. Considering the uncertainties in performance at radiological facilities, HEPA filters for use in dry conditions in the DOE complex have an age limit of 10 years from the date of manufacture (DOE 2003). This lifetime was based on an analysis of multiple HEPA filter research studies on tensile strength degradation. MSRs may implement a similar approach or define objective performance criteria to evaluate the proposed filter designs on a case-by-case graded approach before being replaced or allowed to remain in operation (Matthew et al. 2022).

7.0 Cryogenic Distillation

7.1 Technology Description

Cryogenic distillation is a potential method for separating noble gases (e.g., Xe, Kr) from the preconditioned MSR off-gas stream by concentrating a target product gas as it passes through a distillation column with a packing material. Cryogenic distillation could be employed by an MSR design to allow for separation of radioactive ^{85}Kr from stable Xe, which would allow the first to be stored in tanks until decay allows for release to the environment per pertinent regulatory requirements. MSRs are expected to generate significant amounts of noble fission gases (dependent on reactor power and burnup), as the salts do not provide confinement as would be in the case of cladding in light water reactor fuel. The MSR design could also implement cryogenic distillation to capture the commercial value of high-purity Xe and ^{85}Kr following separation by charcoal decay beds.

The distillation process is based on the difference of vapor pressures between the off-gas components. For an ideal binary off-gas stream, the enhancement in concentration can be described by the relative volatility, α, which is deduced by using Raoult's law to be the ratio between the vapor pressures (P_{vap}) of the two gas components (Aprile et al. 2017), per Equation (7-1). For example, the relative volatility for krypton relative to xenon at -98°C is thus P_{Kr}/P_{Xe} = 1.89 MPa / 0.175 MPa = 10.8. The relative volatility can also be interpreted as a measure of the probability for krypton to migrate into the gaseous phase at liquid xenon temperatures (Aprile et al. 2017).

$$\alpha=\frac{P_{vap}(high)}{P_{vap}(low)} \qquad \text{Equation (7-1)}$$

Figure 7-1 illustrates the configuration of a typical cryogenic distillation column (also referred to as a tower) with partial reflux, i.e., operating via rectification (Aprile et al. 2017). The illustration uses a reference concept where xenon is assumed as the low P_{vap} constituent and krypton is the high P_{vap} constituent in the process. The column incorporates a packing material, i.e., a series of distillation stages (plates) used to successively reduce the concentration and separate the target product gas from the feed-in stream.

The column contains three primary sections: a feeding section where the off-gas stream is injected, a rectifying section above where the more volatile component is enriched, and the stripping section below the feeding section where the more volatile component is depleted. Starting from the bottom of the column, a reboiler with a heater in the stripping section evaporates the binary mixture into an upward gaseous stream with flow rate, *V'*. As the vapor moves up the column, it interacts with the first stage (Plate 1 in Figure 7-1), upon which the vapor condenses into a liquid phase with a higher concentration of the low P_{vap} constituent and lower concentration of the high P_{vap} constituent per the temperature and tortuosity of the stage material. By repeating this process through multiple stages, the fraction of the low P_{vap} constituent will successively increase. At the top of the column, the vapor is partially liquefied with a re-condenser and fed back to the column with a flow rate, *L*. A small fraction of the gas reaching the re-condenser is also diverted as product gas with a high enrichment of high P_{vap} constituent at a flow rate *D*, while the purified liquid with a high enrichment of low P_{vap} constituent is extracted at the bottom with a flow rate, *B*. During this process, the off-gas stream, injected at the feeding section at a flow rate, *F*, may be added to either the up-streaming vapor, *V*, in the rectifying section or to the down-streaming liquid flow, *L'*.

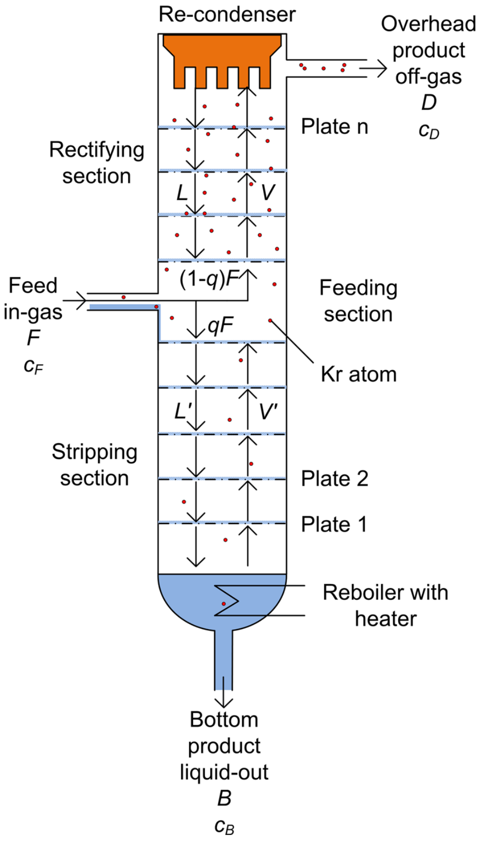


Figure 7-1. Schematic of a distillation column with partial reflux. Volumetric flow rates denoted by capital letters (e.g., *L*, *L'*) and concentrations denoted by lower case letters (e.g., c_F, c_B) (Aprile et al. 2017).

The efficiency of the separation (rectifying) process and achievable DF will depend on the design and operational characteristics of the column, including length of the column, packing material (stage material, number of stages, distance between stages), temperature and pressure gradient along the column, flow rates, boil-up ratio and reflux ratio (i.e., residence time and number of recirculations). The boil-up ratio refers to the vapor fraction partially liquefied at the re-condenser and fed back down the column with a flow rate, *L*, to the liquefied fraction sent for product collection at a flow rate *D*. The reflux ratio refers to the fraction of condensed liquid vaporized up the column at a flow rate, *V'*, to the fraction retained and extracted at the reboiler with a flow rate, *B*. These two parameters control the residence time of each species along the column. The operational parameters will also depend on the relative volatility, α, per the vapor pressures of the constituents of the feed-in gas, which is dependent on the temperature gradient along the column.

As shown in Table 7-1, the differences in boiling points for xenon and krypton are sufficient to allow for a column design where xenon and potential carrier gases (e.g., N_2, Ar) remain in the liquid phase while ^{85}Kr is vaporized in the column. The operating temperature will depend on the pressure inside the column, as higher pressures increase the boiling point of xenon and increases the boil-up ratio (and rectification efficiency). Column designs operate with re-condenser temperatures in the range of -93 to -98°C with a very small gradient to the re-boiler

(1 to 2°C higher)(Wang et al. 2014; Aprile et al. 2017). Operating temperatures will also depend on the packing material design.

During distillation, carrier helium gas would pass through the column with the krypton gas. Multiple columns could be used in series where subsequent columns would be used to rectify high-purity xenon product from the first column by separating the carrier gas. High-purity carrier gas could then be recirculated back to the first column for reuse. Computer modeling with standard chemical process simulation software (e.g., Aspen Plus®, Chemstations™ CHEMCAD) could be used to design the column and define operational parameters per the target DF for an MSR off-gas treatment.

Table 7-1. Boiling and melting points of potential off-gas feed-in species (carriers, target, impurities) for cryogenic distillation (Yusa et al. 1977).

	Gas	Boiling Point (°C)	Melting Point (°C)
Carrier	He	-268.9	-272.2
	N_2	-195.8	-209.9
Target	Kr	-152.3	-156.6
	Xe	-107.1	-111.9
Potential Impurities	CO_2	-78.5	-56.6
	O_2	-183.0	-218.4
	H_2	-252.5	-259.1
	H_2O	100	0

Notes: See Table 1-1. The properties of tritiated impurities will be minimally different than protiated impurities. Carbon bearing impurities could be generated from ^{14}C.

Preconditioning of the MSR feed-in gas is an important consideration for efficient separation by cryogenic distillation (see Figure 1-1). The preconditioning makes sure particulates and off-gas constituents with low vapor pressures are removed (e.g., CO_2, H_2O), that would otherwise condense in the column or generate byproduct gases from radiolysis reactions (e.g., O_3 generation from O_2)(Andrews et al. 2021; Banerjee 2015).

The carrier gas primarily used in non-radioactive industrial applications is N_2, which acts as both cooling and liquid medium in the rectifying section of the column and allows for efficient separations of the rectified product (Riley et al. 2020; Baetsle and Broothaerts 1977; Andrews et al. 2021). N_2 may not be appropriate for use as an MSR cover gas because the N can activate to C-14. N_2 has also been shown to cause nitriding and degradation of mechanical properties of stainless steels used as structural materials (Burak and Chen 2024). After preconditioning, the composition of an MSR off-gas feed-in gas to an initial distillation column would be expected to include three primary constituents, i.e., a carrier gas, xenon and ^{85}Kr. For MSR off-gas feed-in gases, a suitable carrier gas could instead be inert argon, which would prevent the generation of byproduct gas contaminants from radiolysis reactions of N_2 carrier gas and residual oxygen in the off-gas stream, which may condense and potentially plug the packing material (e.g., NO_2) and cause a pressure buildup within the column. Additionally, argon, can activate to Ar-41.

Figure 7-2 illustrates a representative example of structured packing for laboratory-scale cryogenic distillation columns which relies on a wavy design to increase surface area for vapor interaction and heat transfer. The material used for these packaging structures is usually a stainless-steel alloy, which structural stability will not be impacted by beta decay of ^{85}Kr within the column. The number of stages for a distillation column will be based off the target DFs for ^{85}Kr, the specific packing design structure and operational characteristics of the column (e.g., feed-in gas composition, temperature and pressure gradient along the column, flow rates, boil-up ratio and reflux ratio—residence time, number of recirculations), which may vary for different MSR designs. The number of distillation stages may be calculated by the McCabe-Thiele method for low concentration-separation, whereas the volume of packing material needed can be estimated from the height equivalent for one theoretical distillation stage (height equivalent to a theoretical plate [HETP] value) multiplied by the number of distillation stages (Rosendahl et al. 2014).

Figure 7-2. Sulzer Type EX packing used for Xe/Kr cryogenic distillation (Rosendahl et al. 2014).

Cooling of the column could be achieved with mechanical cooling pumps instead of liquid N_2. The latter approach would limit the scale and size of the column and would require reduced volumetric flow rates and longer residence times within the column. The scale of industrial-scale cryogenic columns cooled by liquid N_2 are in the range of 1 to 3 m (diameter) and 10–20 m (height). However, when considering the flow rate conceptualized for the off-gas stream from the MSBR (i.e., 312 L/min)(Andrews et al. 2021), a significantly smaller distillation column of 1/10 industrial scale could be envisioned for an MSR design.

7.2 Safety Considerations

7.2.1 Normal Operations

Performance monitoring of the distillation column is necessary to ascertain that the operation is within the target DF range, trend concentrations in the feed-in gas stream and that the other operational parameters remain within design ranges (e.g., pressures, temperatures, reboiler liquid level). Online process monitoring may be conducted through in-situ spectroscopic techniques, residual gas analyzers or mass spectrometry, pressure transducers, thermocouples, and readouts of liquid level of the reboiler. Sensor and/or sampling locations will be specific to a system design. Monitoring of operational parameter transients from shutdown/restart operations during a batch process (instead of steady-state) may need particular consideration. Section 4 of McFarlane et al. 2020 discusses analytical methods that could be implemented. Amount of liquid at the bottom of the column could be used to quantify how much is removed.

Monitoring will also be important to mitigate occupational safety risks related to the low-temperature, pressurized components, and the gas and liquid products themselves (cryogenic fluid burns, asphyxiation, etc.). These hazards are minimized using protocols developed and practiced industrially (AIGA 2013). However, some key differences include the relatively high radioactivity which creates both dose and heat in the system, which is usually not experienced in industrial applications. Nuclear safety monitoring would also include dose measurements to rule out releases/leaks and confirm adequate system functionality. It is expected that pressure and temperature sensors available to the commercial nuclear industry can be utilized for monitoring MSR off-gas treatment.

The distillation columns have no moving parts and may be designed to accommodate off-gas management for the life of the reactor without changeout of the column packing material. Operator intervention is expected to be limited for a distillation column operating in steady-state. Valves regulating and directing fluid flow within the process may need periodic replacement. Other auxiliary equipment (e.g., cooling pumps, systems for managing large quantities of liquid N_2) may also require maintenance and potential replacement.

7.2.2 Off-Normal Conditions

Table 7-2 provides a partial set of potential off-normal conditions and potential options to address these conditions. The design of the distillation system may incorporate mitigative features to account for these potential conditions. These may include:

- The insulation of the column could be chosen to maximize the thermal inertia for a condition of power loss or failure of system components (e.g., cooling system pump/compressor or other auxiliary components). This would limit gas expansion of the column contents over time and provide time for implementation of a potential option.
- Pressure relief valves to allow for depressurization of the liquid N_2 cooling loop or potentially the off-gas in the column. Off-gas releases could be routed to a redundant power-passive scrubber train.
- The reboiler could also be designed to route liquid overflow to a secondary reservoir to prevent system upset.
- Valves directing the fluid flow could corrode, clog and fail. The design may divert upstream gas flow to a secondary delay system to allow for valve replacement.

Preconditioning of the feed-in off-gas will be necessary to prevent solid particulates from plating or eroding the packing material and high P_{vap} species (e.g., CO_2) from freezing inside the column as these would reduce DFs at sufficient quantities. Removal of long-lived radioiodine compounds from the upstream gas with sorbent materials would prevent binding to the column packing material. Removal of fluorinated and chlorinated species from the upstream gas by the molten hydroxide scrubber may need to be monitored due to potential of corrosion of column internals. The decay process of ^{85}Kr processed through cryogenic distillation will generate stable solid product ^{85}Rb. Rb is known for its role in enhancing corrosion of steel in nuclear material storage (Asmussen and Neeway 2020); therefore, the potential to degrade the column containment over its life may need to be assessed during the design process (e.g., selection of appropriate alloys). A distillation column cooled by liquid N_2 would have a minimal pressure gradient along its length at a nominal operating temperature of -196.1°C. Indications of a significant pressure gradient may be indicative of plated or frozen contaminants. A hazards analysis for the proposed cryogenic distillation system would identify consequences and risks for potential off-normal conditions of the specific design.

Table 7-2. Summary of potential off-normal conditions, potential options, and associated timelines for cryogenic distillation.

Off-Normal Condition	Potential Options
Loss of electrical power leading to a pressure buildup.	Switch to backup electrical power source or generator; Divert upstream gas to redundant power-passive scrubber train (e.g., solid sorbent bed) or delay tanks.
Depressurization of liquid N_2 cooling loop/loss of cooling leading to a pressure buildup. Consideration would be given to occupational risks of ambient O_2 deprivation and/or cryogenic burns.	System shutdown, venting of N_2 via monitored stack; Divert upstream gas to redundant power-passive scrubber train (e.g., solid sorbent bed) or delay tanks.
Column containment failure leading to gas leak or release.	System shutdown; Divert upstream gas to redundant power-passive scrubber train (e.g., solid sorbent bed) or delay tanks; Release via vent stack.
Failure of upstream preconditioning leading to lower separation efficiency.	System shutdown; Divert upstream gas to secondary scrubber train (e.g., solid sorbent bed or molten hydroxide scrubber).

7.3 Considerations on Waste Products

Purified radioactive krypton (^{85}Kr) could be stored in cylinders with solid sorbent materials to reduce storage pressure and immobilize decay products (^{85}Rb). Rb is known for its role in enhancing corrosion of steel in nuclear material storage (Asmussen and Neeway 2020); therefore, the potential to degrade the column containment over its life may need to be assessed. After sufficient decay storage, ^{85}Kr would have sufficiently decayed for environmental release or further separations for commercial reuse. 40 CFR 190, “Environmental Standards for the Uranium Fuel Cycle”, allows for the release of ^{85}Kr to the general environment, although the applicability of the radioactivity limit (see 40 CFR 190.10) may need to be assessed for MSR off-gas operations. Alternative approaches have been developed such as solidification in hot-pressed zeolites and deposition on metals and silicon carbide. If solidified, the materials can be disposed rather than stored.

7.4 Technology Status

7.4.1 Prior Experience

Cryogenic distillation has been commercially deployed for air separation on an industrial scale. The method has also been implemented at nuclear facilities, including the Idaho Chemical Processing Plant (United States), the AZUR plant (Germany), and the Japanese Cryogenic plant (IAEA 1980). Although the process was not implemented in the MSRE, it was proposed for separation of ^{3}H from the off-gas stream for the MSBR concept (Shapiro and Reed 1970).

The Idaho Chemical Processing Plant, a DOE spent fuel reprocessing facility, was the first to implement a process for separation of Kr/Xe using a cryogenic system (Figure 7-3). The off-gas stream was first conditioned by heating, which reduced nitrous oxide to nitrogen and formed water from the reaction of hydrogen and oxygen. The effluent then passed through a condenser, demister, drier, and finally into the cryogenic system, consisting of two cold traps, a distillation unit and a batch still. Once the effluent was cooled in one of the two cold traps, it was heated before entering into the distillation column. In the column, liquid N_2 was introduced, condensing and absorbing higher boiling components (e.g., O_2, Xe, Kr, and Ar). Waste gas (primarily N_2) was discharged while the absorbed gases were further concentrated by re-boiling at the bottom of the column. The concentrated gases were periodically transferred to a batch still where N_2 and O_2 were distilled off. The process was known to generate O_3, which was monitored to avoid ignition hazards for concentrations exceeding two (2) percent (IAEA 1980). This concern does not appear pertinent to MSR operations due to the lack of oxygen in the feed-in off-gas to a cryogenic distillation system. Point Beach Nuclear Plant, Units 1 and 2 installed a cryogenic distillation system in their off-gas system, although it was never operated (NMC 2004).

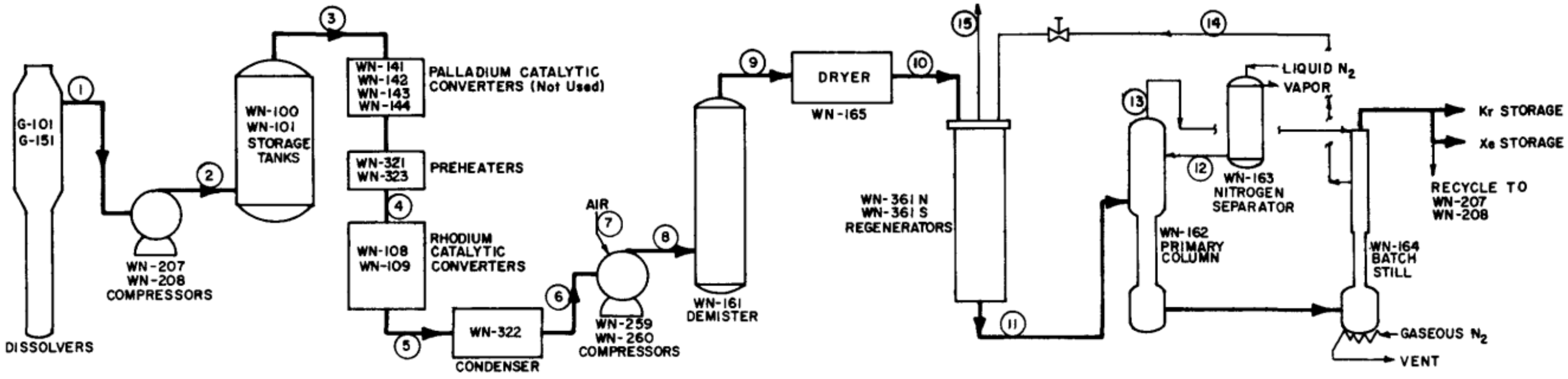


Figure 7-3. Flowsheet for recovery of krypton and xenon from hydrogen-lean dissolver off-gas (Offutt and Bendixsen 1969).

7.4.2 Information Gaps or Needs

Cryogenic distillation is a mature technology practiced in industrial applications, although complex and energy-intensive. The technique could be implemented in MSRs as either a steady-state or batch process, and can be very efficient at rectifying noble gases with modern columns achieving Kr/Xe concentrations in the parts per quadrillion level (1 ppq = 10^{-15} mol/mol) (Aprile et al. 2017). For reference, commercially available high-purity xenon has a krypton concentration of 10 ppb (10^{-9} mol/mol) to 1 ppm (10^{-6} mol/mol) (Rosendhal et al. 2014). Packing structures for column stages are commercially available (Sulzer 2024), although may need qualification for use under radiological conditions pertinent to the MSR off-gas stream (i.e., per MSR concentrations of ^{85}Kr).

8.0 Summary of Attributes of Potential Technologies

This report reviews various technologies and materials for potential implementation in the design of an MSR off-gas system and follows a conceptual framework developed by McFarlane et al. (2020). Table 8-1 summarizes attributes of these technologies and materials, including target off-gas stream species, parameters of importance to decontamination factors (DFs), implementation maturity, system component monitoring parameters and approaches, potential off-normal conditions, and waste form management approaches. The off-gas stream composition and operational parameters for an off-gas system will depend on the specific MSR design and operational characteristics (e.g., fuel salt composition, power level, neutron energy spectrum, fuel burnup). The sequence of components and operations for an off-gas system and its tie to the MSR cover gas will also vary across designs (e.g., tied to fuel salt loop design or available surge cover gas volume). Furthermore, off-normal conditions listed in Table 8-1 may or may not be applicable to a specific MSR off-gas system design. A hazards analysis would support identifying consequences and risks for the full range of off-normal conditions of a specific design, as well as potential actions to return the system to normal operations.

A molten hydroxide scrubber would provide for potential removal of a wide set of off-gas constituents prior to processing via sorbent traps, delay systems, particle traps and/or cryogenic distillation. The scrubber would use a mixture of molten alkaline hydroxides with low boiling points to capture insoluble particles, convert and dissolve salt soluble species, and neutralize and capture acidic species (e.g., CO_2, SO_2, HCl, HF, SeO_2, and TeO_2). Non-condensable gases will pass through the scrubber (e.g., O_2, Ar, He, Kr, and Xe). Conceptually there are other off-gas constituents that could be captured by the molten hydroxide scrubber system (e.g., tritium, Cl_2, F_2, Br_2, I_2, Cs, Sr, Y, La, Se, TeF_4, and SeF_4). Areas that need further development for MSR implementation include optimization of scrubber operational parameters, fluid flow rate versus gas flow rate, online monitoring, systems integration, and routine maintenance requirements (e.g., flushing and replacement of the hydroxide). The effects of radiolysis on the hydroxide eutectic stability have not been investigated. Potential off-normal conditions include loss of heater power (hydroxide bed solidification, over pressurization), bed saturation (lower DFs) and plugging (pressure drops downstream). The ability to switch to a redundant hydroxide scrubber or temporarily divert flow to a passive solid sorbent bed may address the need for draining and flushing of utilized hydroxide and different off-normal conditions.

Solid sorbents have been implemented in nuclear processes for many decades. Charcoal sorbent beds were previously implemented in the Molten Salt Reactor Experiment, although limited testing may be required for implementation in modern reactor designs. Implementation of these materials may be considered of relatively low operational complexity since sorbent beds have no moving parts. However, bed performance will require consideration of factors such as the need for maintaining a specified temperature range (most important for physisorption-based sorbents) and sorption competition with other off-gas species. Potential off-normal conditions include bed saturation, radiolytic damage or fouling/plugging (lower DFs), as well as potential corrosion of the bed containment (loss of captured species). These conditions may be addressed by incorporation of redundant capability (e.g., secondary beds) into the design.

Delay systems involve capture and holdup of short-lived noble fission gases using a combination of solid sorbent beds (delay beds or tanks) or multi-chamber pathways (e.g., 10-min delay pipelines). Delay systems have been implemented in medical isotope production facilities as well as operating light water reactors. A delay system could be implemented either as a first off-gas system component to allow very-short lived radionuclide decay (e.g., prior to the molten hydroxide scrubber) and/or as an emergency drain tank for mitigation of a potential off-normal condition. Potential off-normal conditions relate to sorbent material performance, as stated above, as well as potential increases in off-gas stream flow rates (e.g., due to power level changes in reactor), particularly for delay lines without sorbent material, which would reduce DFs. If the design reflects the ability to switch to a redundant set of delay beds or tanks, this could address different off-normal conditions.

Particle traps may be used to remove aerosols, salt mists and splashes, particulates, and condensed daughters of noble gases. High-efficiency particulate absorbing (HEPA) filters have been widely deployed in the nuclear industry. Potential off-normal conditions relate to filter plugging, improper emplacement, or rips and tears decreasing capture efficiency. These conditions may be address by off-gas diversion to a secondary filter train.

Cryogenic distillation could be employed by an MSR design to allow for separation of radioactive ^{85}Kr from stable Xe, which would allow the ^{85}Kr to be stored in tanks until decay allows for release to the environment per pertinent regulatory requirements. Although this technology is considered to be mature, other factors including the presence of low-temperature pressurized components as well as the need for maintaining cooling introduce moderate challenges for implementation. The upstream gas would be adequately scrubbed of H_2O and other condensable species that would decrease DFs. Potential off-normal conditions include failure of upstream gas conditioning, loss of electrical power (loss of cooling, active gas flow) leading to pressure buildup within the column, depressurization of the liquid N_2 and loss of cooling, and failure of the column containment leading to gas release. These conditions may be addressed by back-up electrical power sources and diversion of upstream gas to power-passive delay systems.

It is noted that this report does not make any conclusions about the conformance of any specific technology or design with regulations that would apply to MSR off-gas systems.

Table 8-1. Summary of attributes of potential technologies.

	Molten Hydroxide Scrubber	Solid Sorbents	Delay Off-Gas System	Particle Traps	Cryogenic Distillation
Target Species	Particles, mists, aerosols, reactive volatile species (e.g., CO_2, SO_2, SeO_2, TeO_2, HF, HCl, Cl_2, F_2, Br_2, I_2, Cs, Sr, Y, La, Se TeF_4, SeF_4)	Water species (e.g., ^{3}HHO or $^{3}H_2O$) Iodine and iodide species Noble gases (e.g., ^{139}Xe, ^{137}Xe, ^{135m}Xe, ^{135}Xe, ^{133m}Xe, ^{133}Xe, ^{89}Kr, ^{87}Kr)	Noble gases (e.g., ^{139}Xe, ^{137}Xe, ^{135m}Xe, ^{135}Xe, ^{133m}Xe, ^{133}Xe, ^{89}Kr, ^{87}Kr)	Aerosols, salt mists and splashes, particulates, and condensed daughters of noble gases	Noble gases (e.g., Xe, ^{85}Kr, Ar)
Implementation Maturity	Low The concept of a molten hydroxide scrubber has only been studied on a laboratory scale.	High Solid sorbents have been implemented in nuclear processes for many decades.	High Delay systems have been implemented in medical isotope production facilities as well as boiling water reactors.	High High-efficiency particulate absorbing (HEPA) filters have been widely deployed in the nuclear industry.	High Cryogenic distillation has been implemented in industrial applications as well as nuclear facilities.
Monitoring/Control Parameters	Monitoring to ascertain that the bed is functioning properly, is at correct temperature, and parameters are within design targets.	Monitoring of the decontamination factor for target analyte concentrations.	Monitoring of performance to ascertain that delay system is functioning properly.	Monitoring of system to ascertain differential pressure is in design-specific range and to monitor temperature and relative humidity.	Monitoring of the decontamination factor and other operational parameters.
Monitoring/Control Instrumentation	Monitoring would require instrumentation for alarm function, equipment control and data collection. Sensors used for: • temperature (e.g., thermocouples), • pressure, • corrosion/erosion (e.g., ultrasonic), • flow (e.g., flow meter), • radiation monitoring, and • turbidity.	Monitoring of target analytes would be done through: • spectroscopy (e.g., optical, laser-induced breakdown, mass), • radiation detection methods, and • temperature.	Monitoring would include: • thermocouples for temperature monitoring, • mass spectrometer for *in situ* detection, and • pressure monitoring for potential leakage.	Monitoring instrumentation would include: • pressure gauge, and • thermocouples for temperature monitoring.	Monitoring instrumentation would include: • thermocouples for temperature monitoring, • pressure gauge, • level sensor, and • spectroscopy.

	Molten Hydroxide Scrubber	**Solid Sorbents**	**Delay Off-Gas System**	**Particle Traps**	**Cryogenic Distillation**
Off-Normal Conditions	• Loss of heater power • Over pressurization • System is saturated • Corrosion of components • Instrumentation or control failure • Piping plug or high pressure drops • Loss of hydroxide recirculation	• High saturation • Radiolytic damage • Plugging/fouling • Corrosion of the bed	• Unexpected spike in off-gas stream flow rate • High saturation • Radiolytic damage • Plugging/fouling • Corrosion of the bed	• Improper emplacement • Plugged filters • Rip or tear within filter media	• Loss of electrical power • Depressurization or loss of cooling • Containment failure; off-gas leak or release
Waste Forms/Processes	• Treated hydroxide salt • Process as a mineral or glass waste form	• Waste may be stored in compressed gas or high-integrity containers. • Disposal by hot pressing or consolidation (e.g., grout).	• Delay beds can be designed for the life of the facility. • Replacement may be needed to maintain treatment efficiency.	• Used filters typically require treatment (e.g., compaction, grouting) prior to disposal. • May be stored onsite prior to offsite treatment and disposal.	• Gases, such as Kr, may be stored with solid sorbents to immobilize reactive decay products until sufficient decay for commercial use. • Treatment of wastes may also include solidification in hot-pressed zeolites and deposition for disposal.

9.0 References

10 CFR Part 20. *Code of Federal Regulations.* Title 10, *Energy,* Part 20 “Standards for Protection Against Radiation.”

10 CFR Part 50. *Code of Federal Regulations.* Title 10, *Energy,* Part 50 “Domestic Licensing of Production and Utilization Facilities.”

40 CFR Part 61. *Code of Federal Regulations.* Title 40, *Protection of the Environment*, Part 190, “Environmental Radiation Protection Standards for Nuclear Power Operation.”

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