Technical Letter Report

[TLR-RES/DE/REB-2026-04]

Aqueous and Pyrochemical Reprocessing Off-Gas and Ventilation Systems: Technical Descriptions to Aid Regulatory Considerations

Date:
February 2026

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INL/RPT-25-85408

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Prepared for the
U.S. Department of Energy
Office of Nuclear Energy
Under DOE Idaho Operations Office
Contract DE-AC07-05ID14517

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EXECUTIVE SUMMARY

This report was written to aid the U.S. Nuclear Regulatory Commission (NRC) in decision-making regarding future updates and/or development of new regulatory guides (RGs) for nuclear fuel reprocessing plant off-gas and ventilation systems. Volatile radionuclides (i.e., certain fission products) are released into off-gas streams during the mechanical and chemical operations of reprocessing. The topic of off-gas and ventilation systems is important for the control of these radionuclides for reasons of occupational and environmental health and safety. This report provides technical descriptions of topics related to nuclear fuel reprocessing, review of prior RGs, considerations for new RGs, and a bibliography for additional technical resources. Since the issuance of the prior RGs (ca. 1975), aqueous reprocessing has made significant advancements by offering more separations options to meet the needs of diverse fuel cycle options and safeguard concerns. Pyrochemical reprocessing, as it is envisioned now, has not been demonstrated commercially.

The primary purpose of nuclear fuel reprocessing is to recover fissile materials from spent fuel. Most often, select actinides are to be retained and all other elements rejected. Aqueous reprocessing relies on separations between aqueous and organic phases. Tailored separations are achieved by control of the aqueous redox chemistry and selection of organic extractants. Pyrochemical reprocessing relies on separations between molten salts and electrochemically reduced actinide metals. Tailored separations are achieved by control of molten salt redox chemistry and selection of reductive processes. In either case, fresh fuel is made from the actinides recovered and purified from spent fuel.

During reprocessing operations, radionuclides become volatile by three pathways: (1) they are noble gases like Kr-85, (2) they combine with other elements to form gases like I-129, and (3) during high-temperature operations, they form compounds with high vapor pressures like Cs-137. Regardless of the cause of the volatility, radionuclides must be managed by the off-gas and ventilation systems to prevent their release into the atmosphere.

There are both similarities and differences in the behavior and management of volatile radionuclides between aqueous and pyrochemical reprocessing plants. Voloxidation is an optional head-end[a] technique that has not yet been demonstrated commercially. It is the high-temperature roasting of oxide fuel,

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[a] The term head-end refers to the initial mechanical, chemical, or thermal steps in a nuclear fuel reprocessing plant that prepares spent fuel for materials recovery operations.

which can be applied to both aqueous and pyrochemical reprocessing. If applied, the bulk of volatile radionuclides can be released during voloxidation; however, this is highly dependent on the conditions of the voloxidation process. Otherwise, in aqueous reprocessing, the bulk are released during decladding and fuel dissolution, and in pyrochemical reprocessing, the bulk are released during decladding and oxide reduction or electrorefining. Differences between aqueous and pyrochemical reprocessing are summarized in the following table.

Table ES1. Aqueous reprocessing and pyrochemical reprocessing.

<table>
<tr><th>Aqueous Reprocessing</th><th>Pyrochemical Reprocessing</th></tr>
<tr><td>Mature technology that has been demonstrated at industrial scales.</td><td>Early technology that has been demonstrated at research and engineering scales.</td></tr>
<tr><td>Off-gas capture technologies applied and demonstrated (in other countries) to meet emissions regulations.</td><td>Off-gas capture technologies have not been applied and demonstrated to meet emissions regulations.</td></tr>
<tr><td colspan="2">Voloxidation may be used as a head-end process in both flowsheets. If applied, the majority of the volatile fission products will be released during decladding and voloxidation.</td></tr>
<tr><td>Majority of volatile fission products are released during cladding breeching and fuel dissolution.</td><td>Majority of volatile fission products are released during cladding breeching, oxide reduction, and electrorefining.</td></tr>
<tr><td>Off-gas includes vapors of water, acidic solutions, and organic solvents that condense upon cooling and require wet scrubbing.</td><td>Off-gas includes volatile metal oxides and metal chlorides that condense upon cooling and require filtration.</td></tr>
<tr><td>Iodine is released to the off-gas.</td><td>Iodine is captured in the chloride salts, but the extent of capture has not been validated as sufficient to meet emissions requirements.</td></tr>
<tr><td>Carbon is released to the off-gas.</td><td>Carbon is captured in the chloride salts, but the extent of capture has not been validated as sufficient to meet emissions requirements.</td></tr>
<tr><td>Tritium follows the aqueous solutions and organic solvents and is released into the off-gas. Tritium spreads throughout the entire process as tritiated water.</td><td>Tritium is released to the off-gas with varied chemical speciation.</td></tr>
<tr><td colspan="2">Noble gas fission products such as krypton-85 and xenon are released to the off-gas.</td></tr>
</table>

The report assesses the changes in regulatory framework since the RGs were issued. Title 40 of the *Code of Federal Regulations* (40 CFR) Part 190, which was promulgated in the late 1970s, includes emissions-based limits that take into account the electrical energy produced by the fuel cycle. Both 10 CFR 20 and 40

CFR 190 also include dose-based limits that require licensees to take into account the cumulative dose to the public from multiple radionuclides arising from both liquid discharges and gaseous emissions. Therefore, the most restrictive emissions standards for a particular reprocessing technology will be determined by factors such as the plant capacity and location.

The contents of RGs 3.20 and 3.32 can be reduced to a few high-level functional capabilities listed below that cover the intent of the guides, which hinges on system reliability and safety, and are broadly applicable to both aqueous reprocessing and pyrochemical reprocessing technologies. Ventilation and off-gas systems best practices include:

- Direct facility air flow from the areas of lower contamination potential toward the areas of higher contamination potential.
- Maintain safe functionality during anticipated abnormal conditions, such as process upsets, equipment malfunctions, losses of power, fires, severe weather, flooding, earthquakes, etc.
- Be equipped with process monitors that alarm when abnormal conditions occur.
- Provide means for process sampling.
- Be constructed of materials that are heat-resistant and nonflammable and be equipped with smoke and fire suppression systems.
- Have isolation devices that prevent the flow of contamination into uncontaminated areas during operations, inspections, maintenance, and abnormal conditions.
- Designed such that components can be maintained, repaired, decontaminated, and replaced as needed.
- Accommodate anticipated equipment and process changes.
- Withstand the influx of materials such as process gases, dust, fumes, and condensates.
- Withstand the deleterious effects of radiation and provide adequate radiation containment and shielding for worker safety.
- Prevent the spread of contamination into unwanted areas and the environment.
- Meet the standards of safety significant and safety class as identified by Federal guidance and regulations.

There appears to be no impediment to the RGs being used by a potential vendor for an aqueous or pyrochemical reprocessing facility, as the overall guidance is general enough to be considered technology neutral. The NRC could consider consolidation of the two RGs into a single RG that broadly contains the high-level functional capabilities listed above in addition to others that are yet to be determined. Specific functional capabilities bordering on engineering design requirements may be appropriate when design specifications for the next proposed reprocessing facility are known.

The development of more explicit guidance may be delayed until more is understood about how reprocessing will be pursued. In the interim, it is recommended that designers and reviews consider the general high level functional capabilities noted in the previous paragraph as well as the DOE documents presented in section 12.1.1 of this report to the extent they are relevant.

Potential approaches to managing off-gases and volatile radionuclides are diverse and implementation scenarios will have wide variance based on the specific flowsheet designs. Specific capture requirements for each volatile radionuclide will be impacted by such choices as aqueous reprocessing versus pyrochemical reprocessing, flowsheet selection, facility design, and facility siting. Technical descriptions of technologies deployed currently in reprocessing facilities are included in this report to help inform future guidance considerations, namely the ventilation systems at the Idaho National Laboratory Fuel Conditioning Facility and the Hot Fuels Examination Facility.

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FOREWORD

Three complementary reports were prepared for the Nuclear Regulatory Commission:

- *Aqueous and Pyrochemical Reprocessing Off-Gas and Ventilation Systems: Technical Descriptions to Aid Regulatory Considerations*, INL/RPT-25-85408
- *Topics Related to Pyrochemical Reprocessing: Technical Descriptions to Aid Regulatory Considerations*, INL/RPT-25-85431
- *Aqueous and Pyrochemical Reprocessing Chemical Safety: Technical Descriptions to Aid Regulatory Considerations*, INL/RPT-25-87767.

The first report describes key issues associated with volatile radionuclides during both aqueous and pyrochemical reprocessing and provides a technical basis for any future NRC guidance pertaining to off-gas and ventilation systems at a reprocessing facility. The key topics include the chemistry of release into off-gas ventilation systems, the capture from off-gas ventilation systems, Federal regulations governing the release to the atmosphere, technical descriptions of existing hot cell facilities, and an extensive bibliography on related publications.

The second report expands the technical descriptions of pyrochemical reprocessing into subjects beyond the reference case discussed in the first report. These subjects include the application of pyrochemical reprocessing to various fuel types, the unit operations of voloxidation and oxide reduction, the technology of fluoride volatility, salt waste management, and the consequences of aged fuels. The report identifies expected safety-related aspects of existing potential technologies to support potential near-term reprocessing applications.

The third report provides a deeper assessment of key chemical process safety issues associated with both aqueous and pyrochemical reprocessing. These key issues include chemical excursions, pyrophoric materials, and chemical safety.

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FIGURES

TABLES

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ACRONYMS

ADAMS	Agencywide Documents Access and Management System
ALARA	As low as reasonably achievable
ANL	Argonne National Laboratory
ASU	Air separation unit
CAM	Continuous air monitor
CFR	Code of Federal Regulations
COG	Cell off-gas
DBP	Dibutyl phosphate
DOE	Department of Energy
DOG	Dissolver off-gas
DU	Depleted uranium
EPA	Environmental Protection Agency
FCF	Fuel Conditioning Facility
HEU	High-enriched uranium
HFEF	Hot Fuel Examination Facility
HLW	High Level Waste
HTGR	High-temperature gas-cooled reactors
IAEA	International Atomic Energy Agency
IFR	Integral Fast Reactor
INL	Idaho National Laboratory
IROFS	Item relied on for safety
ISA	Integrated safety analysis
JFCS	Joint Fuel Cycle Study
LCC	Liquid cadmium cathode
LEU	Low-enriched uranium
LLW	Low-level waste
LWR	Light-water reactor
MFC	Materials and Fuels Complex
MOF	Metal-organic framework
MOG	Melter off-gas
MOX	Mixed oxide
MSR	Molten salt reactor
MSRE	Molten Salt Reactor Experiment

NESHAP	National Emissions Standards for Hazardous Air Pollutants
NRC	Nuclear Regulatory Commission
ORNL	Oak Ridge National Laboratory
PUREX	Plutonium Uranium Reduction Extraction
RCRA	Resource Conservation and Recovery Act
RG	Regulatory guides
SFR	Sodium-cooled fast reactors
SFT	Spent Fuel Treatment
SNF	Spent nuclear fuel
TBP	Tributyl phosphate
THORP	Thermal Oxide Reprocessing Plant
TREAT	Transient Reactor Test Facility
TRISO	Tristructural isotropic
TRU	Transuranic
VOG	Vessel off-gas
WOG	Waste off-gas

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Aqueous and Pyrochemical Reprocessing Off-Gas and Ventilation Systems

Technical Descriptions to Aid Regulatory Considerations

1. INTRODUCTION

The purpose of this report is to present information that will aid the U.S. Nuclear Regulatory Commission (NRC) in decision-making regarding future updates and/or development of new regulatory guides (RGs) for nuclear fuel reprocessing plant off-gas and ventilation systems. The previous RGs addressing these systems were written over 50 years ago, as follows:

- RG 3.20, "Process Off-Gas Systems for Fuel Reprocessing Plants," U. S. Atomic Energy Commission, Directorate of Regulatory Standards, February 1974
- RG 3.32, "General Design Guide for Ventilation Systems for Fuel Reprocessing Plants (for Comment)," U.S. Nuclear Regulatory Commission, Office of Standards Development, September 1975.

The year 1976, around the time these RGs were issued, saw the beginnings of a substantial U.S. policy shift that effectively ended the pursuit of commercial reprocessing. Soon afterward, all existing and planned projects were canceled.

Despite the lack of commercial implementation, research into reprocessing has continued. The preceding 50 years have seen the development of new aqueous reprocessing technologies and the introduction of pyrochemical reprocessing, which did not exist in its current form at the time RG 3.20 and RG 3.32 were published. The advances in aqueous reprocessing have provided many new options for chemical separations to meet the needs of different fuel cycle options and safeguard requirements. Pyrochemical reprocessing as it is currently pursued can be applied to oxide fuels and metallic fuels to recover actinides as metals. Both approaches are now subject to additional regulations that post-date RG 3.20 and RG 3.32, with the most impactful arising from the Environmental Protection Agency (EPA) (i.e., 40 CFR 190). This evolving landscape prompts this report, which allows for the examination of RGs 3.20 and 3.32 in the context of both technological advancement and regulatory shifts.

The primary focus of RGs for reprocessing plant off-gas and ventilation systems is to address the inherent release of volatile radionuclides into the off-gas streams from the mechanical and chemical operations of reprocessing. The off-gas streams that bear radionuclides must be contained in engineered systems until the radionuclides can be captured and removed. Limits on the atmospheric discharge of radionuclides are controlled by the *Code of Federal Regulations* (CFR), as follows:

- 40 CFR 190, "Environmental Radiation Protection Requirements for Normal Operations of Activities in the Uranium Fuel Cycle, Volume I," U.S. Environmental Protection Agency, November 1976
- 10 CFR 20, "Standards for Protection Against Radiation," Nuclear Regulatory Commission, Up to Date.

This report covers a variety of subjects related to nuclear fuel reprocessing. There are four major sections. Sections 2 to 9 provide technical descriptions of aqueous and pyrochemical reprocessing. Sections 10 to 11 examine relevant RGs and Federal regulations. Recommendations for the development of future RGs are provided in Section 12. Section 13 provides technical descriptions of the ventilation systems of two hot cell facilities at the Idaho National Laboratory (INL). Section 14 contains a bibliography of documents relevant to this subject matter.

2. VOLATILE FISSION PRODUCTS

Volatile fission products are isotopes derived from the constituents of spent fuels that have the propensity to report to a gas phase during reprocessing and waste processing operations. These operations occur at both ambient temperatures and high temperatures. Volatile fission products include stable isotopes and radioisotopes. Stable isotopes can be hazardous due to their chemical properties, while radioisotopes can be hazardous due to their chemical and radiochemical properties. Volatility heightens safety concerns because gases can be challenging to contain and highly mobile once released to the ambient atmosphere.

All nuclear fuels undergo fission in a similar manner. While in the reactor core, fuels undergo the processes of fission, transmutation, and radioactive decay. Once removed from the core, fission and transmutation essentially stop while radioactive decay continues. The thermal energy released during radioactive decay is called decay heat. The amount of decay heat, or the decay heat load, is measured in units of energy per time per mass, or watts per mass. Here *mass* can be some measure of the mass of the spent fuel, but it must be defined. The decay heat load decreases approximately exponentially as a function of the cooldown period, which is the time since fission processes were stopped. Often this is equivalent to approximately the time the fuel has been out of the reactor core.

Factors such as fuel composition, reactor core neutronics, burnup, and cooldown have significant impacts on the properties and compositions of spent fuels. There are also significant differences between spent fuel types related to the compositions of the matrices containing the fission products and fissile metals. These matrices include graphite, ceramics, alloys, and molten salts of many different compositions and forms.

Most nuclear reactors use fuels that are contained in metal cladding. The spent fuels are removed from the reactor and allowed a cooldown period prior to reprocessing. From a reprocessing perspective, metal-clad spent fuel is merely a material from which fissile metals are to be recovered and purified for use in making new fuel. The fissile metals are retained in the fuel cycle, and the fission products are rejected from the fuel cycle. This basic concept applies to any type of solid fuel that is removed from the reactor core, not just metal-clad fuels.

Molten salt reactors (MSRs) with fuel salts are different. In this case, there is an inventory of fuel salt that is continuously circulating through the reactor core. Some concepts for managing fuel salt chemistry rely on diverting a slip stream of the fuel salt away from the reactor and into a chemical processing plant for treatment. In this scenario, there is no cooldown period. Treatment can be used for the recovery and purification of fissile metals to make fresh fuel salts or treatment to recover and dispose of fission products to increase the operational life of the fuel salt. In either case, the slip stream that was diverted away from the reactor is returned to the reactor in a purified form to keep the reactor operating. However, these scenarios are supposition as the on-line treatment of MSR fuel salts has yet to be demonstrated. Alternatively, treatment of the fuel salt may be deferred until the end-of-life of the reactor.

Chemical reagents utilized in aqueous reprocessing are subject to damaging effects caused by decay heat and radiolysis. Therefore, spent fuels must be allowed sufficient cooldown before aqueous reprocessing commences. In contrast, the chemical processes used in pyrochemical reprocessing are not subject to the damaging effects caused by decay heat and radiolysis. However, it is still necessary to manage the decay heat either by providing sufficient cooldown before reprocessing (as is done with aqueous reprocessing) or by managing the radioisotopes responsible for producing a decay heat load as they accumulate in the process salts. Decay heat management is just as important to pyrochemical reprocessing as it is to aqueous reprocessing. The only differences are in the strategies used to manage the decay heat.

Table 1 is a list of isotopes that are associated with volatile species during reprocessing and waste processing operations. This list comes from a survey of technical literature and considers MSR operations. The table contains the isotope name, its half-life, and notes on its source. In Table 1, half-lives shorter than 1 year are shaded gray and are generally not of concern in significantly aged spent fuels. In general, chemical properties are determined by the behavior of the element, whereas radiochemical properties are determined by the behavior of the isotope. Therefore, the properties associated with *volatility* are essentially dependent on the element and independent of the isotope, particularly with the heavier elements and particularly with respect to issues related to off-gas ventilation systems.

An important consideration is the half-lives of the isotopes and the age of the spent fuels. For example, if the spent fuel has been out of the reactor for 5 years, there is no need to be concerned with short-lived fission product isotopes whose half-lives are measured in terms of a few days—because for all practical purposes, those short-lived isotopes will be gone. However, there are exceptions. For example, if the short-lived isotope is a daughter product of radioactive decay of a parent nuclide, then it may be present for the duration of the decay of that parent and is produced by mechanisms other than fission. Another example is MSR fuel salts. For example, the continuous treatment of fuel salts, as described earlier, represents a source of fresh fuel. Therefore, the short-lived isotopes that are not of concern during conventional reprocessing activities of aged spent fuels are of concern during the treatment of MSR fuel salts.

Table 1 includes helium, argon, krypton, xenon, and radon, which are noble gases. The table also includes hydrogen, fluorine, chlorine, and iodine, which are gases in their elemental states under ambient atmospheric conditions. These elements are highly chemically reactive and form numerous compounds with other elements, many of which are gases or have high vapor pressures. The other elements in Table 1 are not as commonly recognized for their volatility, and in their elemental states, they are not particularly volatile. But they can form volatile species with other elements under certain conditions encountered during reprocessing and waste processing operations. Some of the metals in Table 1 are Resource Conservation and Recovery Act (RCRA) metals. Specific chemistries are discussed later in the report. The table is meant to summarize the radioisotopes that may be of concern during reprocessing operations. The radioisotopes that will be of concern during a specific application are dependent on the characteristics of the spent fuel and the reprocessing technologies to be applied.

Table 1. Summary of volatile and semivolatile, nuclides and radionuclides.

Isotope	Half-Life	Source
Hydrogen-3 Tritium	12.3 y	Neutron activation of deuterium (H-2) Neutron activation of lithium (Li-6 and Li-7) Ternary fission
Helium-3	Stable	Alpha decay product
Helium-4	Stable	Alpha decay product
Carbon-14	5.73E3 y	Neutron activation of nitrogen (N-14 and N-15) Neutron activation of oxygen (O-16 and O-17) Neutron activation of carbon (C-13)
Fluorine-18	109 min	Proton activation of oxygen (O-18)
Sodium-22	2.60 y	Proton activation of neon (Ne-22)
Sodium-24	14.95 hr	Neutron activation of sodium (Na-23)
Chlorine-36	3.01E5 y	Neutron activation of chlorine (Cl-35) Cl-35 is an impurity in graphite
Argon-37	35.0 d	Neutron activation of calcium (Ca-40)
Argon-39	268 y	Neutron activation of potassium (K-39)

Isotope	Half-Life	Source
Argon-41	110 min	Neutron activation of argon (Ar-40)
Selenium-74-76-77-78-80	Stable	RCRA metal
Selenium-79	3.27E5 y	Binary fission
Bromine-79-81	Stable	Binary fission
Krypton-85	10.76 y	Binary fission
Krypton-85m	4.48 hr	Binary fission
Krypton-87	76.3 min	Binary fission
Krypton-88	2.83 hr	Binary fission
Strontium-89	50.6 d	Binary fission
Stontium-90	28.9 y	Binary fission
Strontium-91	9.63 hr	Binary fission
Strontium-92	2.66 hr	Binary fission
Molybdenum-99	65.9 h	Binary fission Neutron activation of molybdenum (Mo-98)
Technetium-99	2.11E5 y	Binary fission Beta decay of molybdenum (Mo-99)
Ruthenium-103	39.26 d	Binary fission
Ruthenium-105	4.44 hr	Binary fission
Ruthenium-106	373.6 d	Binary fission
Rhodium-103	Stable	Binary fission Beta decay of ruthenium (Rh-103)
Rhodium-103m	56.1 min	Binary fission
Rhodium-105	35.3 hr	Binary fission
Rhodium-106	30.0 s	Binary fission Beta decay of ruthenium (Rh-106)
Rhodium-106m	131 min	Binary fission Beta decay of ruthenium (Rh-106)
Palladium-106	Stable	Binary fission Decay of rhodium (Rh-106 and Rh-106m)
Silver-107-109	Stable	Binary fission RCRA metal
Silver-109m	39.8 s	Binary fission
Silver-111	7.43 d	Binary fission
Silver-113	5.37 hr	Binary fission
Cadmium-106-108-110-111-112-114	Stable	Binary fission RCRA metal
Cadmium-113m	14.1 y	Binary fission

Isotope	Half-Life	Source
Cadmium-115	53.5 hr	Binary fission
Tin-121m	43.9 y	Binary fission
Antimony-125	2.76 y	Binary fission
Antimony-127	3.85 d	Binary fission
Antimony-129	4.37 hr	Binary fission
Tellurium-127	9.35 hr	Binary fission
Tellurium-127m	109 d	Binary fission
Tellurium-129	69.6 min	Binary fission
Tellurium-129m	33.6 d	Binary fission
Tellurium-131	25.0 min	Binary fission
Tellurium-131m	30 hr	Binary fission
Tellurium-132	3.20 d	Binary fission
Iodine-129	1.61E7 y	Binary fission Neutron activation of xenon (Xe-129)
Iodine-131	8.02 d	Binary fission Neutron activation of tellurium (Te-130)
Iodine-132	2.30 hr	Binary fission
Iodine-133	20.8 hr	Binary fission
Iodine-134	52.5 min	Binary fission
Iodine-135	6.58 hr	Binary fission
Xenon-133	5.25 d	Binary fission
Xenon-135	9.12 hr	Binary fission
Xenon-135m	15.3 min	Binary fission
Cesium-134	2.06 y	Binary fission
Cesium-135	1.33E6 y	Binary fission
Cesium-136	13.2 d	Binary fission
Cesium-137	30.2 y	Binary fission
Barium-134-135-136-137-138	Stable	Binary fission RCRA metal
Barium-137m	2.55 min	Binary fission
Barium-139	82.3 min	Binary fission
Barium-140	12.8 d	Binary fission
Radon-220	55.6 s	Decay series of thorium (Th-232)
Radon-222	3.82 d	Decay series of uranium (U-238)

2.1. Vapor Pressure

The radioisotopes listed in Table 1 can report to the gas phase by three predominant mechanisms: (1) the fission product can itself be a gas at ambient temperatures, (2) the fission product can be included in a compound that is a gas at ambient temperatures, and (3) the fission product can report to the gas phase as a consequence of vapor pressure effects at high temperatures. Understanding the concept of vapor pressure is key to understanding the behavior of semivolatile fission products. The three common states of matter are solid, liquid, and gas. Vapor pressure forms in the gas phase.

From chemical thermodynamics, when a solid is placed in an evacuated sealed chamber, a gas phase will form such that the solid and gas are in equilibrium with each other. (Here we are ignoring any contributions from the chamber itself.) The composition of the gas is dependent on the composition of the solid and the temperature of the system. The same is true for any liquid and any combination of solid and liquid. It is also possible that the solid or liquid is entirely converted to a gas, leaving behind no solid or liquid. This illustrates the concept of vapor pressure. In other words, all solids and all liquids have associated vapor pressures. However, these vapor pressures may or may not be of practical concern. Below are two examples to illustrate the nature of vapor pressure.

Example One

> The vapor pressure of carbon gas surrounding a piece of graphite under ambient atmospheric conditions is of no particular concern. However, use that same graphite as an electrode in an arc lamp and the situation changes. The extremely high temperatures encountered in an arc lamp are sufficient to vaporize and ionize the carbon. This illustrates the importance of considering temperature when discussing vapor pressures. Again, consider carbon. If carbon is in the form of carbon dioxide or methane, then the carbon is in the gas phase under ambient atmospheric conditions. This illustrates the importance of speciation. The phrase "of practical concern" was used above. This phrase implies that when considering volatile species, there is a threshold of vapor pressure below which there is no concern and above which there is a concern. These thresholds are often based on considerations of health and safety or process chemistry.

Example Two

> A closed vessel is completely evacuated except that its volume is about half filled with pure water (H_2O). The temperature remains constant at 25°C. Several things will immediately and simultaneously begin to happen as follows:
>
> 1. Some of the water will evaporate filling the empty volume of the vessel with $H_2O(g)$
> 2. Some of the liquid water will dissociate to form hydronium cations (H_3O+) (i.e., formed when a proton attaches to a water molecule) and hydroxide anions (OH-)
> 3. Some of the water vapor will dissociate to form hydrogen gas ($H_2(g)$) and oxygen gas ($O_2(g)$).
>
> Of course, the predominant component in the liquid phase is $H_2O(l)$, and the predominant component in the gas phase is $H_2O(g)$. But other species do exist in both phases as consequences of the thermodynamic and kinetic properties of a system comprised of hydrogen and oxygen at a given temperature, pressure, and volume.

> The total absolute pressure of the gas phase that develops over the liquid phase is the vapor pressure of water at 25°C. If the temperature is increased slightly, some of the liquid water will evaporate and the vapor pressure will increase. If the temperature is decreased slightly, some of the water vapor will condense and the vapor pressure will decrease. If the volume of the vessel is increased slightly, some of the liquid water will evaporate to fill the volume and the vapor pressure will remain constant. If the volume of the vessel is decreased slightly, some of the water vapor will condense and the vapor pressure will remain constant. These behaviors are examples of classical physical chemistry effects.

All the radioisotopes listed in Table 1 exhibit some form of vapor pressure behavior that will be influenced by temperature and the speciation of the condensed phase (liquid or solid) and the vapor phase. It is maybe counterintuitive, but thermodynamics indicate that all substances have a vapor pressure. That vapor pressure may be so slight as to be completely inconsequential, but nevertheless it exists. As discussed above, vapor pressure increases with temperature. Therefore, some of the radioisotopes listed in Table 1 are only a concern at high temperatures because at ambient temperatures, their vapor pressures are so low as to be inconsequential.

3. BREACHING CLADDING AND DECLADDING

Cladding is the barrier between the fuel and the coolant. Barring mechanical failure, cladding is the containment that prevents fuels and fission products from escaping into the coolant during reactor operations. The material properties of the cladding must be compatible with the fuel on one side and the coolant on the other side. Therefore, there are different cladding materials for use with different reactor types and fuel types. For example, oxide fuels for light-water reactors (LWRs) are commonly clad in zirconium alloys, whereas oxide and metallic fuels for sodium-cooled fast reactors (SFR) are commonly clad in stainless steels. Tristructural isotropic (TRISO) fuels for high-temperature gas-cooled reactors (HTGRs) and fluoride salt-cooled high-temperature reactors offer a unique example of cladding that consists of alternating spherical layers of silicon carbide and pyrolytic graphite surrounding small particles of what is referred to as fuel kernels of uranium oxide or uranium oxycarbide. These TRISO particles are then embedded in graphite or ceramic compacts. Oxide, hydride, and metallic fuels for research reactors are commonly clad in aluminum. Cladding is not applicable to fuel salts that are circulating through MSRs.

For aqueous and pyrochemical reprocessing alike, at some point, the cladding is breached to expose the fuels to the process chemicals. Cladding is breached during operations such as chopping, shredding, dissolution, and decladding.

Decladding is the operation of separating the cladding from the fuel; there are different methods of decladding. Mechanical decladding is the physical separation of cladding from the fuel, and chemical decladding is the chemical dissolution of the cladding from the fuel without chemically altering the fuel. However, decladding may or may not be applied as a stand-alone unit operation. In some reprocessing schemes, the fuel and cladding are dissolved together in the nitric acid dissolution vessel. In other cases, the fuel is dissolved leaving most of the cladding undissolved. Yet in other cases, the cladding is dissolved leaving most of the fuel undissolved. What becomes of the cladding is determined by the reprocessing strategy.

There are many variants of reactors, fuels, decladding techniques, and reprocessing strategies. A decision to incorporate decladding as part of the reprocessing strategy for either aqueous or pyrochemical reprocessing can be based on several factors. As an example, decladding can reduce the quantity of liquid waste generated during aqueous reprocessing. A few examples are given below illustrating how cladding can be managed during reprocessing.

Mechanical Decladding of LWR Oxide Fuel with Zirconium Alloy Cladding and SFR Oxide Fuel with Stainless Steel Cladding

The engineering challenges for mechanical decladding are similar between LWR oxide fuels clad in zirconium alloys and SFR oxide fuels clad in stainless steels. The two greatest challenges may be (1) managing fine particles that form as the brittle oxide fuels are fractured and (2) achieving adequate separation and recovery of the oxide fuels from the claddings. Metallurgical interactions between the fuel and the cladding cause the fuel to stick to the cladding, making it challenging to effectively separate the two from each other. The extent of disengaging the fuel from the cladding can be improved by applying thermal oxidation (voloxidation) to convert the UO_2 to U_3O_8. Once separations are achieved, the fuel is advanced to reprocessing, and the cladding is advanced to a separate waste treatment process.

Chemical Decladding of LWR Oxide Fuel with Zirconium Alloy Cladding

Chemical decladding also presents engineering challenges. One approach to chemical decladding is the chlorination of the zirconium alloy cladding by gaseous compounds such as chlorine ($Cl_2(g)$), hydrogen chloride ($HCl(g)$), and carbon tetrachloride ($CCl_4(g)$). The chlorination of zirconium metal by $Cl_2(g)$ to form zirconium chloride ($ZrCl_4(g)$) is shown in the following equation:

$$Zr + 2\,Cl_2(g) = ZrCl_4(g).$$

Zirconium chloride has a high vapor pressure making it a volatile salt species. The volatility of $ZrCl_4$ is used to separate and transport the $ZrCl_4$ away from the fuel and any cladding residuals while it is in the gas phase. It can then be condensed into a receiver vessel and handled as solid $ZrCl_4$. This chlorination approach has been proposed not only as a means of decladding but also as a means of recycling the zirconium. The volatile zirconium chloride can be captured and converted back to zirconium metal to make new cladding, as the oxide fuel is advanced to reprocessing. This form of chemical decladding is proposed for oxide fuels that are clad in zirconium alloys but not for oxide fuels that are clad in stainless steels.

Zirconium cladding does not dissolve well in concentrated nitric acid solutions due to the formation of a passivating layer of zirconium oxide (ZrO_2). To overcome this problem, a mixture of nitric acid (HNO_3) and hydrofluoric acid (HF) is used according to the following reaction:

$$3\,Zr + 13\,HNO_3 + 3\,HF = 3\,ZrF(NO_3)_3 + 4\,NO + 8\,H_2O.$$

Chemical Decladding of SFR Oxide Fuel with Stainless Steel Cladding

An historic approach to the chemical decladding of oxide fuels clad in stainless steel is an aqueous dissolution process that dissolves the stainless steel but not the oxide fuel. An example is dissolving the stainless steel cladding in a sulfuric acid solution, followed by dissolving the oxide fuel in a nitric acid solution.

Chemical Decladding of Research Reactor (RR Fuels with Aluminum Cladding)

There are many types of aluminum clad RR fuels. For example, the uranium fuel can be present as uranium oxide, uranium alloys (with aluminum, molybdenum, and zirconium), and uranium hydride. Methods for removing the aluminum cladding include chlorination to form volatile aluminum chloride. The volatile $AlCl_3$ (often present as an Al_2Cl_6 adduct) can be transported and recovered in a similar manner as was described for $ZrCl_4$ (above).

$$2\,Al + 3\,Cl_2(g) = 2\,AlCl_3(g)$$

Another approach to chemical decladding is the dissolution of aluminum cladding in a caustic solution. The aluminum is dissolved leaving behind the oxide or metal fuel as shown in the following reaction:

$$2\,Al + 2\,NaOH + 2\,H_2O = 2\,NaAlO_2 + 3\,H_2(g).$$

Chopping of SFR Metallic Fuel with Stainless Steel Cladding

During pyrochemical reprocessing of SFR metallic fuel with stainless steel cladding, the fuel is chopped into small pieces to breach the cladding and expose the fuel to the process chemicals. In this approach, the chopped fuel is loaded into an anode basket. Uranium electrorefining is performed in a molten salt at high temperature (e.g., 500°C) to recover the impure uranium in the anode as purified uranium on the cathode. The stainless steel cladding remains unaffected in the anode basket and is advanced to a separate treatment process.

4. AQUEOUS REPROCESSING

Plutonium Uranium Reduction Extraction (PUREX) Process was developed in the 1940s at the Metallurgical Laboratory at the University of Chicago, which became the Argonne National Laboratory in 1946. The PUREX process was first used at the Savannah River Site and then the Hanford Site for the recovery of uranium and plutonium from spent nuclear fuels from dedicated plutonium production reactors. This plutonium was used for military applications. The PUREX process is also suited for reprocessing nuclear fuels from commercial reactors generating electrical power.

This section provides a description of a generalized PUREX flowsheet capable of recovering product streams of purified uranium and plutonium. Identifying a specific type of spent fuel for this discussion is not necessary because with sufficient head-end processing, the PUREX process is suited for a variety of fuel types including oxides and metals. In addition to uranium and plutonium, the PUREX process can be adapted to recover product streams of purified neptunium, americium, and curium, as well as select fission products. In most cases, a generalized PUREX flowsheet is a sufficient starting point for analysis of volatile species distribution. Off-gas changes between various aqueous reprocessing flowsheets are not hugely significant, unless voloxidation is introduced on the front end. What follows are descriptions of the process chemistry and process flowsheet of a classic PUREX process.

4.1. PUREX Process Chemistry

Irradiated fuel is dissolved in a concentrated (approximately 9 molar) nitric acid (HNO_3) solution. The dissolution process releases volatile fission product gases from the fuel and generates nitrogen oxide (NO_x) and hydrogen (H_2) gases. The dissolution process is exothermic and produces substantial heat. Depending on the fuel type, the dissolution process for the cladding may be a separate unit operation from the dissolution process for the fuel. Fuel dissolution dissolves both the actinides and the fission products.

The key to the PUREX process is manipulating the redox states of uranium and plutonium, such that they selectively partition between aqueous and organic solutions in several sequential extraction cycles. In this example, the organic solution is tributyl phosphate (TBP: $(CH_3CH_2CH_2CH_2O)_3PO$) in a kerosene (approximate range of hydrocarbons from $C_{12}H_{26}$ to $C_{15}H_{32}$) diluent.

The plutonium and uranium in the dissolver solution are present as uranyl (VI) cations (UO_2^{2+} + $2NO_3^-$) and plutonyl (VI) cations ($PuO_2^{2+} + 2NO_3^-$). Efficient extraction of the plutonium requires reducing plutonyl (VI) cations to plutonium (IV) cations ($Pu^{4+} + 4NO_3^-$). To accomplish this reduction, nitrite anions (NO_2^-) are created in the solution either by the addition of sodium nitrite salt ($NaNO_2$) or by sparging nitrogen oxide (NO_x) gases. The nitrite anions rapidly reduce the plutonium valence from (VI) to (IV).

The uranyl (VI) cations and plutonium (IV) cations form neutral complexes with the nitrate anions present in the dissolver solution. These neutral species are extracted to an organic solution by the action of TBP as $UO_2(NO_3)_2{\bullet}2TBP$ and $Pu(NO_3)_4{\bullet}2TBP$. This extraction cycle partitions the bulk of the uranium and plutonium to the organic solution while leaving the bulk of the fission products in the aqueous solution. Approximately 95% of the fission products are removed from the process in this extraction.

In the next extraction cycle, the plutonium (IV) cations are stripped from the organic solution to an aqueous solution by reducing the plutonium (IV) cations to plutonium (III) cations. Reagents such as ferrous sulfamate ($Fe(H_2NO_3S)_2$), hydroxylamine nitrate (HAN) ($H_4N_2O_4$), or hydrazine (N_2H_4) are used for this reduction. Uranium (IV) nitrate ($U(NO_3)_4$) can also be used as a reductive reagent for plutonium recovery in advanced PUREX flowsheets. In the next extraction cycle, with the plutonium effectively removed, uranium is stripped from the organic solution using dilute nitric acid.

The plutonium in the aqueous solution is present as purified plutonium (III) nitrate ($Pu(NO_3)_3$). Plutonium is precipitated from the solution as plutonium (IV) oxalate ($Pu(C_2O_4)_2{\bullet}6H_2O$) by reaction with oxalic acid dihydrate ($H_2C_2O_4{\bullet}2H_2O$). This precipitate can be calcined to form plutonium oxide (PuO_2).

The uranium in the aqueous solution is present as purified uranyl nitrate ($UO_2(NO_3)_2$). Uranium is precipitated from the solution as ammonium diuranate ($(NH_4)_2U_2O_7$) by reaction with ammonium hydroxide (NH_4OH). This precipitate is typically converted to either UO_3 or U_3O_8, the latter of which is the most thermodynamically stable oxide of uranium. These oxides are, subsequently, converted to uranium (IV) oxide (UO_2) for use in nuclear fuel fabrication. The conversion of UO_3 and U_3O_8 to UO_2 is accomplished by reduction with hydrogen gas (H_2) or a mixture of an inert gas and hydrogen gas. The uranium (IV) oxide and plutonium (IV) oxide (PuO_2) can be blended to make a mixed oxide (MOX) fuel.

4.2. PUREX Flowsheet Description

As outlined above, the key to the PUREX process is successfully manipulating the redox states of uranium and plutonium. In this example, the first cycle extracts uranium and plutonium from the aqueous dissolver solution to an organic solution. The bulk fission products remain in the aqueous dissolver solution, called the first cycle raffinate. In the second cycle, uranium and plutonium are separated from each other by selectively stripping the plutonium from the organic solution to a clean aqueous solution. It should be noted that future commercial demonstrations of aqueous reprocessing would likely not produce a pure plutonium stream due to proliferation concerns. In the third cycle, the uranium remaining in the organic solution is extracted to a clean aqueous solution. Because the separation efficiency of each extraction cycle is not perfect, there are additional extraction stages to improve the separations and recoveries of uranium, plutonium, and process chemicals.

A flowsheet of the off-gas handling systems is shown in Figure 1. In this flowsheet, the spent fuel is chopped or otherwise prepared for dissolution into a nitric acid solution. The majority of the volatile fission products are released during the head-end operations of fuel chopping, decladding (not shown), voloxidation (not shown), and dissolution. Details about decladding, voloxidation, and off-gas fission product capture are discussed in separate sections.

Excess moisture in the off-gas from fuel dissolution is removed in a condenser before the gas proceeds to additional treatments. The solution containing the dissolved fuel is transferred to tankage for input accountancy and then to tankage for process feed. Off-gases from fuel chopping, decladding (not shown), voloxidation (not shown), fuel dissolution, and subsequent tankage all report to the off-gas treatment system. When wet scrubbing is employed for off-gas treatment, the bulk of the water is removed in a condenser before proceeding to further treatment (i.e., radioactive noble gas removal and HEPA, fiberglass, and/or sand filtration) and ultimately released to the atmosphere.

A flowsheet of the *PUREX Extraction Cycles* is shown in Figure 2. Separations are based on the transfer of select dissolved metals between aqueous and organic solutions. The aqueous solution is predominantly nitric acid (HNO_3), which serves as a salting reagent to dissolve the spent fuel. The organic solution is predominately a mixture of kerosene ($C_{12}H_{26}$ to $C_{15}H_{32}$) and dissolved TBP. Kerosene is called the *diluent* and other hydrocarbons can take the place of kerosene.

The dissolved spent nuclear fuel (SNF) enters at the top of the flowsheet in Figure 2. All the rectangular boxes in this figure are mechanical systems that provide intimate mixing, followed by disengagement, of the aqueous and organic solutions with counter-current flow of the solutions. The mechanical systems can be centrifugal contactors, pulse columns, or mixer settlers. The purpose of these systems is to enhance the rate and extent of the chemical transfers between the two immiscible liquid phases.

In the boxes labeled *Diluent Wash*, *Decontamination/Extraction*, and *Decontamination/Scrub*, the organic solution (dash-dot line moving to the right) collects the uranium and plutonium from the aqueous solution (solid line moving to the left). The outgoing aqueous solution reports to *High Level Waste* (HLW) and *Acid Recycle,* shown in Figure 3. This effluent stream can be processed further to separate and collect select metals such as neptunium, minor actinides, and select fission products. Otherwise, the waste metals are processed as HLW. The remaining concentrated acid reports to *Acid Recycle* shown in Figure 3.

The organic stream containing uranium and plutonium flows to the box labeled *U/Pu Partition*. In this operation, the chemistry of the aqueous solution is altered by the addition of nitric acid (HNO_3) and a reductant such as hydroxylamine nitrate (HAN) ($NH_3OH \bullet NO_3$). This chemical change partitions the plutonium into an aqueous solution and leaves the uranium in an organic solution. From here, the plutonium and uranium streams flow to two separate polishing circuits.

The boxes at the bottom of the figure labeled *Diluent Wash* and *Uranium Strip* are the uranium polishing circuit. Here the uranium is transferred from the organic solution to the aqueous solution as uranyl nitrate ($UO_2(NO_3)_2$). The purified uranium solution is collected in the tankage. The outgoing organic solution reports to *Solvent Wash* shown in Figure 4.

The boxes in the middle of the figure labeled *Plutonium Polish/Extraction*, *Plutonium Polish Strip*, and *Diluent Wash* are the plutonium polishing circuit. Here the plutonium is transferred from the aqueous solution into an organic solution, and then back into an aqueous solution. The purified plutonium solution is collected in the tankage. The outgoing organic solution reports to *Solvent Wash* shown in Figure 4. The remaining concentrated acid reports to *Acid Recycle* shown in Figure 3.

There are four *Diluent Wash* units in Figure 2. Their purpose is to recover TBP from the aqueous phase. Diluent is circulated in a closed loop through each of the *Diluent Wash* units until the TBP concentration is too high, rendering the solution ineffective as a wash. The spent saturated diluent is then bled to waste or to *Solvent Wash* shown in Figure 4.

A flowsheet for the acid recycle systems is shown in Figure 3. The purpose of this system is to recover, purify, and reconcentrate the nitric acid. The spent acid accumulates in the tankage that feeds the evaporator. The evaporate is the acid solution, and the bottoms are the fission product HLW solids. The acid solution is next concentrated in the distillation tower. The evaporate is water, and the bottoms are the concentrated acid. Excess concentrated acid reports to a low-level waste (LLW) solids. The recycled concentrated acid is conditioned and reused. The overall process flowsheet has many more than one acid recycle system to accommodate different solution chemistries.

A flowsheet for the solvent wash system is shown in Figure 4. The purpose of this system is to recover and purify both the diluent and TBP. The organic solvent streams accumulate in tankage as feed to the circuit. In the first box labeled *Water Wash*, the solvent is contacted with dilute nitric acid. In the first box labeled Caustic *Wash*, the solvent is contacted with a caustic solution of sodium carbonate (Na_2CO_3). This removes the acidic degradation products of TBP, such as dibutyl phosphate (DBP), monobutyl phosphate (MBP), and phosphoric acid (H_3PO_4). These contaminants ultimately report to a LLW form. In the second box labeled Caustic Wash, the solvent is contacted with sodium hydroxide (NaOH). This removes more of the same contaminants. In the second box labeled Water Wash, the solvent is contacted again with dilute nitric acid. The conditioned solvent is reused.

Figure 1 to Figure 4 were adapted from an earlier report and redrawn for use in this report [1]. Some of the process descriptions are based on information from earlier training materials [2].

4.3. Fission Products and Minor Actinides

In the classic PUREX process described here, uranium and plutonium are recovered as separate purified products, while the fission products and minor actinides are partitioned to the raffinate as waste. Some flowsheet variants, often denoted as “Advanced PUREX” processes, are meant to provide one or more of the following features:

- Co-collection of uranium and plutonium, as to avoid the recovery of a purified plutonium product

- Collection of long-lived minor actinides such as neptunium, americium, and curium for transmutation in fast reactor fuels
- Collection of short-lived fission products such as cesium and strontium for transmutation in fast reactor fuels.

The recovery of long-lived minor actinides and short-lived fission products will reduce the long-term heat load and the radiotoxicity of the raffinate waste [3]. Additional information on the PUREX process, including additional process details, can be found in reference reports [4, 5].

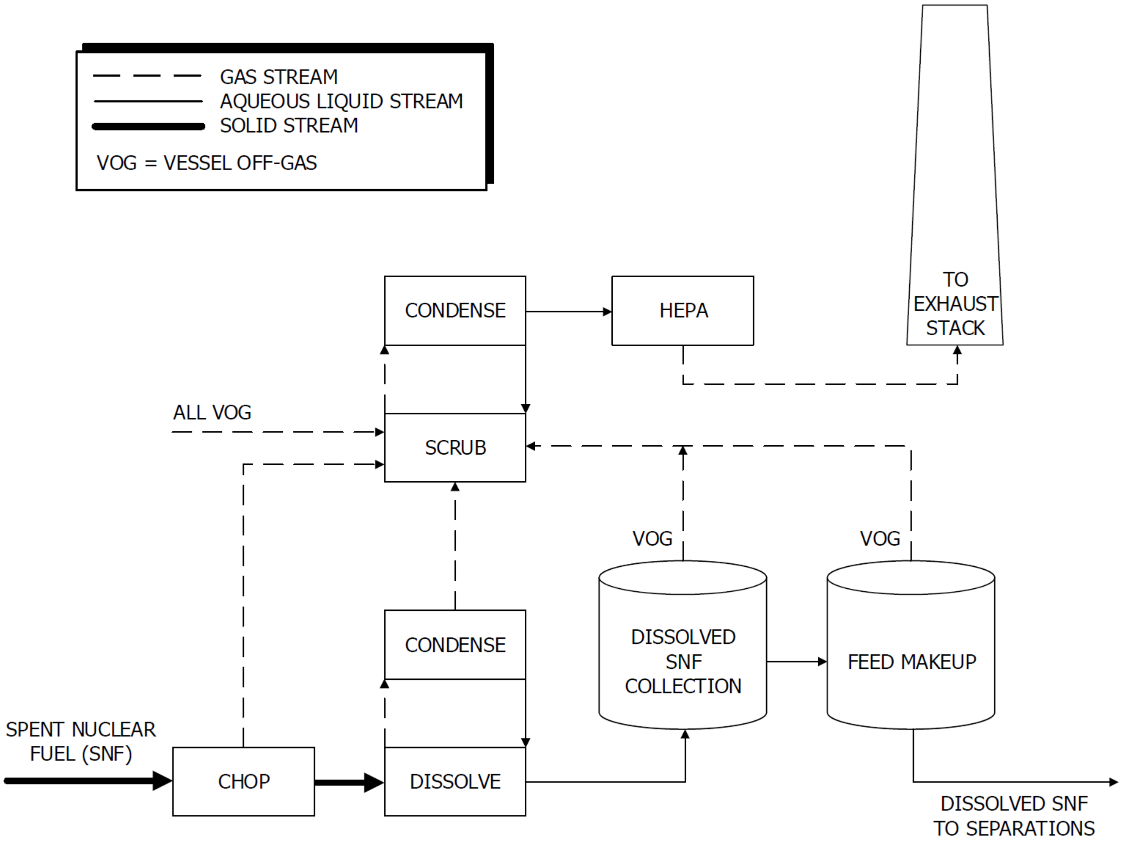

Figure 1. Flowsheet of PUREX off-gas handling systems.

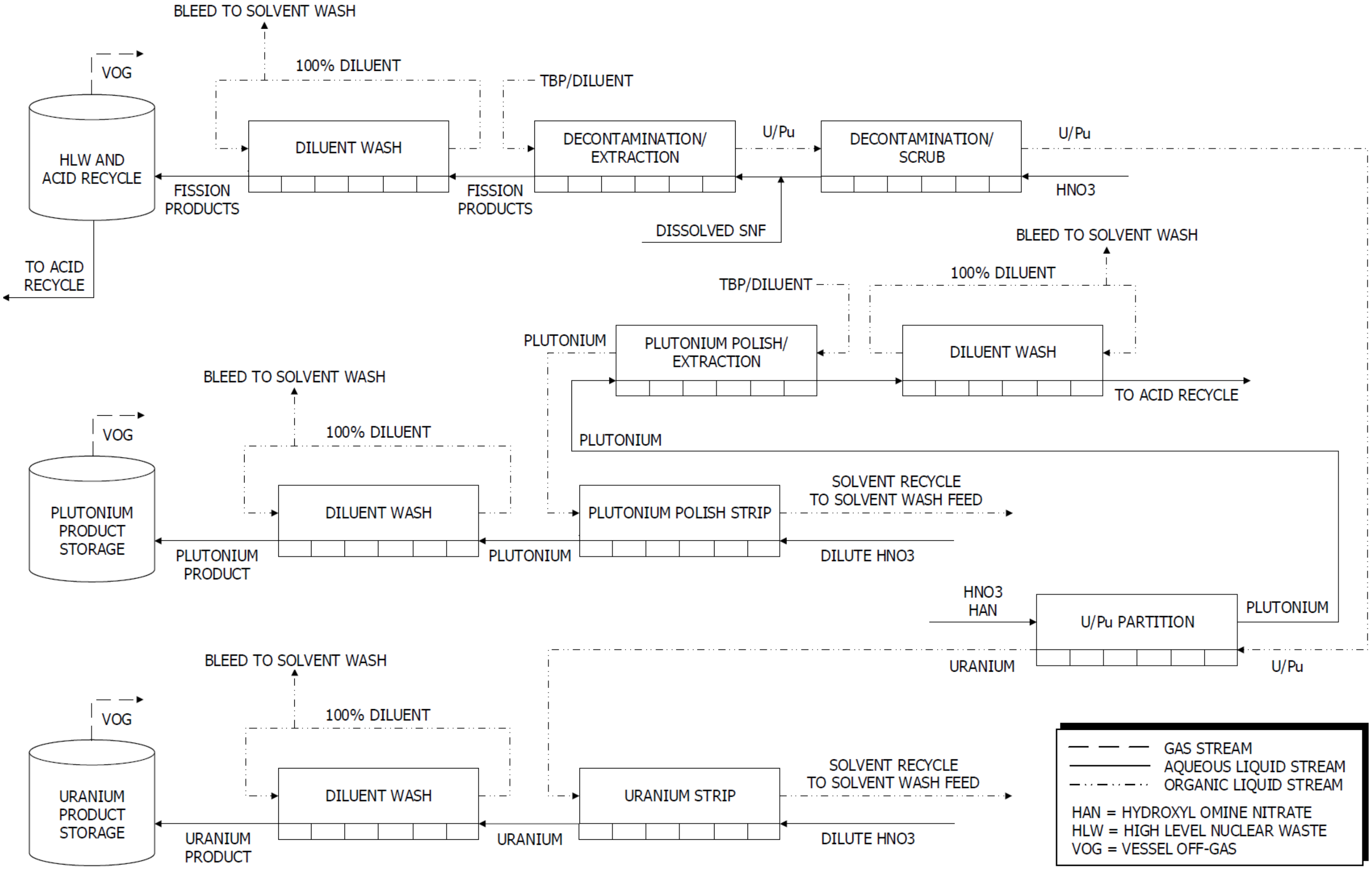

Figure 2. Flowsheet of PUREX extraction cycles.

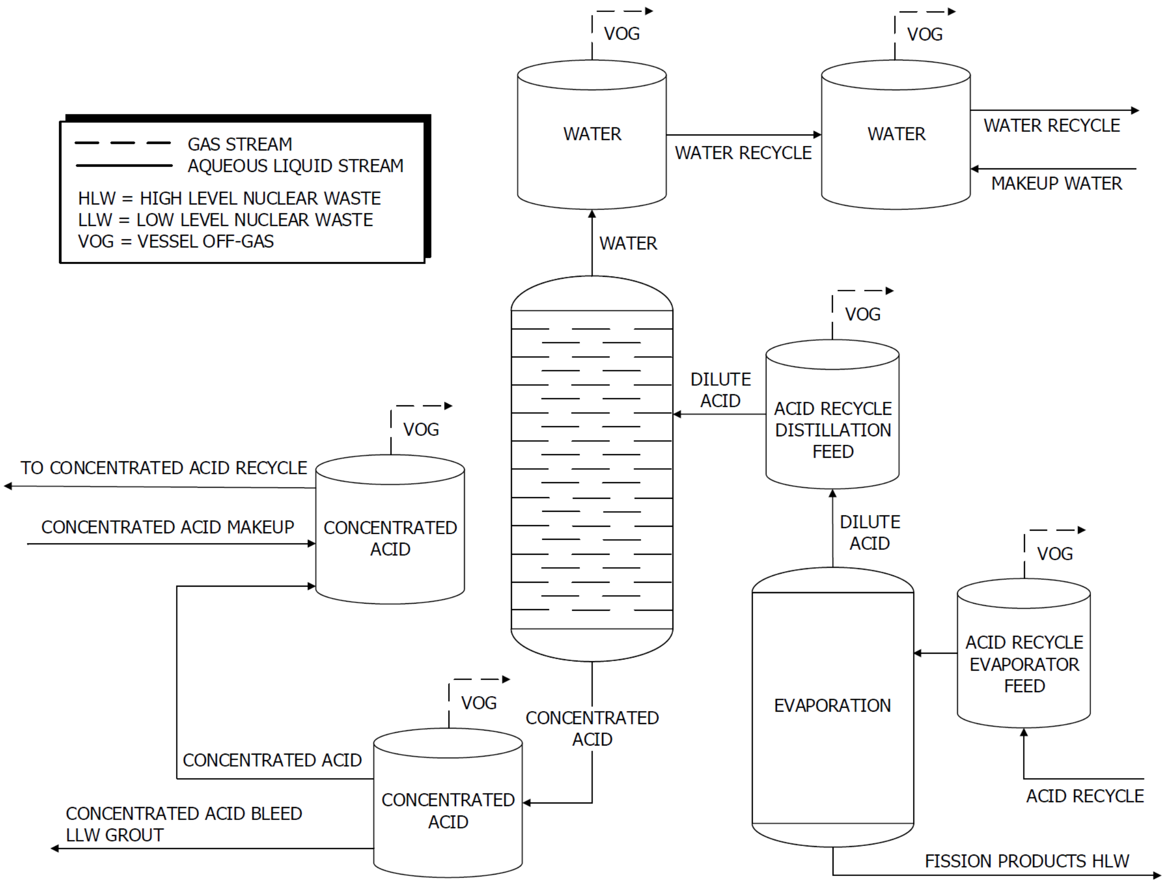

Figure 3. Flowsheet of PUREX acid recycle systems.

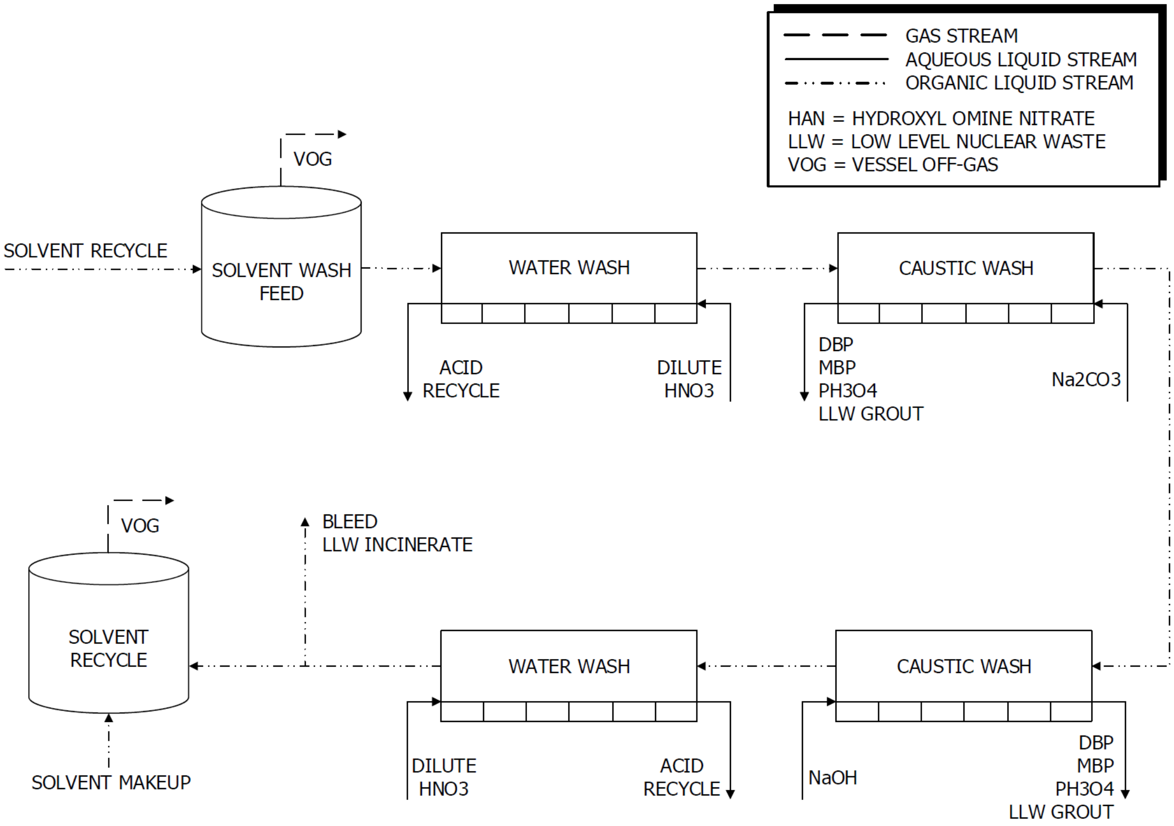

Figure 4. Flowsheet of PUREX solvent wash systems.

4.4. H-Canyon at Savannah River Site

The description of the PUREX process presented in this section was largely taken from materials describing the PUREX flowsheet from the H-Canyon at the Savannah River Site. This PUREX facility was designed to recover weapons grade plutonium from dedicated plutonium production reactors utilizing depleted uranium and natural uranium fuels. The photographs in Figure 5 and Figure 6 show the facility as it looks today and as it looked during construction, respectively. These figures convey the massive size of the facility. The photograph in Figure 7 shows the interior of the radiation-shielded canyon fitted with PUREX processing equipment [6].

Figure 5. Photograph of H-Canyon at Savannah River as it looks in its present condition.

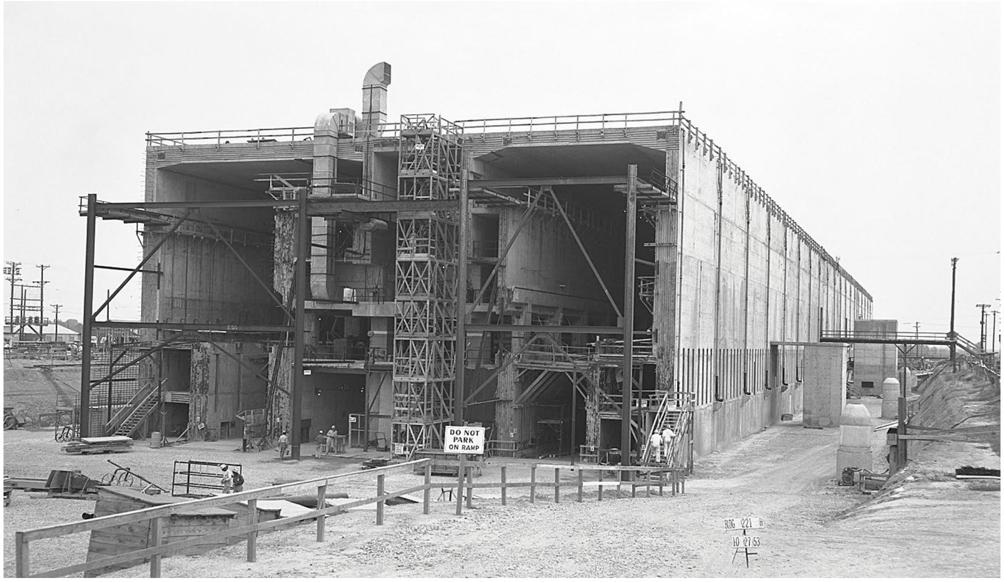

Figure 6. Photograph of H-Canyon as it looked during construction in the 1950s.

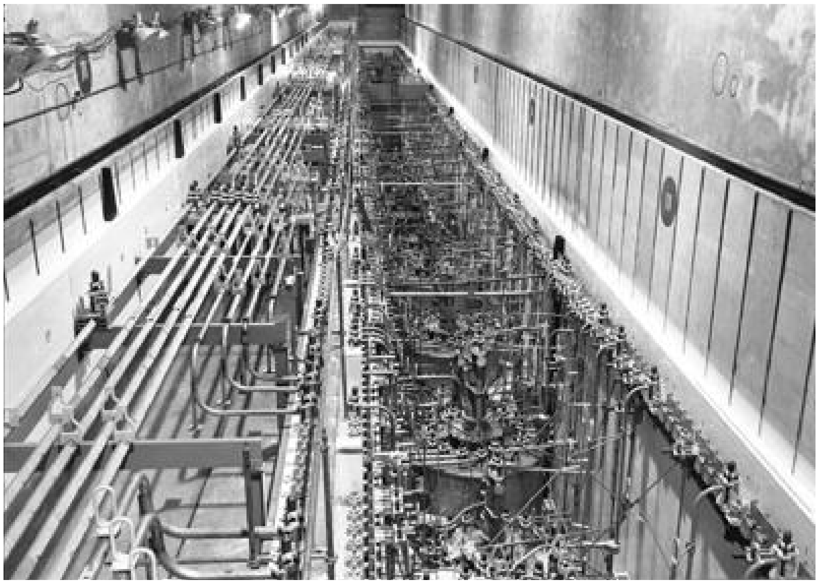

Figure 7. Photograph of the interior of the hot-side of the H-Canyon.

5. PYROCHEMICAL REPROCESSING

The flowsheets of three variants of pyrochemical reprocessing are described here. The first is the reprocessing of Experimental Breeder Reactor-II (EBR-II) driver fuel as envisioned under the Integral Fast Reactor (IFR) Program. EBR-II driver fuel was composed of sodium-bonded high-enriched uranium (HEU) metal alloyed with 10 wt% zirconium (U/10Zr fuel). The purpose of this flowsheet was two-fold: (1) to recover purified uranium to make new binary (U/Zr) metallic fuel and (2) to recover purified uranium and plutonium (and minor actinides) as a uranium/plutonium metal alloy to make new ternary (U/Pu/Zr) metallic fuel.

The second is disposition of EBR-II driver fuel and blanket material under the Spent Fuel Treatment (SFT) Program [7]. The shutdown of EBR-II occurred in 1994 about a year before the IFR Program was able to begin the reprocessing demonstrations. Following the shutdown of EBR-II, the mission changed from the IFR Program to the SFT Program. Because bond-sodium is highly reactive with water, which can lead to the generation of explosive hydrogen gas, untreated EBR-II driver fuel and blanket material did not meet the acceptance requirements for direct disposition to the Yucca Mountain Nuclear Waste Repository.[b] The purpose of this flowsheet was to recover the uranium in the driver fuel and blanket material, convert the bond-sodium into sodium chloride (NaCl), and place the fission products into waste form suitable for disposition. The HEU in the driver fuel was down-blended with depleted uranium (DU) and recovered as purified low-enriched uranium (LEU) ingots, and the uranium in the blanket material was recovered as purified DU ingots. The plutonium and minor actinides were not recovered in metallic forms but were instead allowed to accumulate in the chloride salts along with other fission products.

The third is reprocessing of LWR fuel as demonstrated under the Joint Fuel Cycle Study (JFCS) Program [8]. LWR fuel is composed of LEU uranium oxide (UO_2). The purpose of this flowsheet is to recover purified uranium and plutonium (and minor actinides) as a uranium/plutonium metal alloy to make new ternary (U/Pu/Zr) metallic fuel for SFRs. The spent LWR fuel is merely the source of plutonium that is recovered and used for making SFR ternary metallic fuels.

There is an alternative pyrochemical reprocessing flowsheet developed for mixed oxide (MOX) fuels that will be mentioned briefly. In the United States, it is called the Salt Cycle Process, and in the Russian Federation, it is called the Russian Institute of Atomic Research (RIAR) Process. In this process, the spent MOX fuel is chlorinated into a high-temperature molten salt, and the uranium and plutonium are ultimately recovered as purified UO_2 and PuO_2. This flowsheet has not been pursued in the United States since the 1960s and was described in a separate report [9].

5.1. Process Chemistry of Uranium Electrorefining

The key to pyrochemical reprocessing is the chemical separations, which occur in the uranium electrorefining cell as shown in Figure 8 [7, 10]. The anode is a steel basket that holds the chopped metal fuel, and the cathode is a steel mandrel that collects the electrorefined uranium metal. The anode basket and cathode mandrel are submerged in a salt comprised of the eutectic composition of lithium chloride (44 wt% LiCl) and potassium chloride (56 wt% KCl) with a nominal bulk concentration of uranium trichloride (5 wt% UCl_3).[c] The salt is maintained at 500°C. An external DC power supply is used to pass electrical current between the anode and cathode.

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[b] Development of the Yucca Mountain Nuclear Waste Repository Project was ultimately cancelled in 2010 before it was operational as a repository. The repository never received any nuclear wastes.

[c] The eutectic composition in the specific mixture of LiCl and KCl that has the lowest melting temperature within the range of compositions between 100% LiCl and 100% KCl.

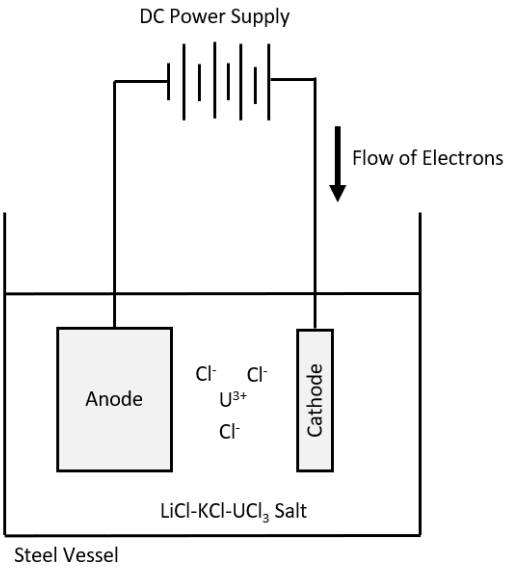

Figure 8. Simple schematic of a uranium electrorefining cell.

The molten salt is an ionic liquid that serves as the electrolyte to facilitate the chemical reactions. A net charge balance is maintained between anions (negative charge species) and cations (positive charge species). At the anode, uranium metal (U^0) is oxidized into the salt as a uranium cation (U^{3+}). The uranium cations migrate through the salt from the anode to the cathode. At the cathode, the uranium cations are reduced back to uranium metal.

Oxidation at the anode:

$$U^0 = U^{3+} + 3\ e^-$$

Reduction at the cathode:

$$U^{3+} + 3\ e^- = U^0$$

Electrical charge flows through the external circuit as the migration of electrons, and through the salt as the migration of cations.

The spent fuel contains a large array of elements created by fission and transmutation processes which are partitioned during electrorefining. Generally, uranium is the most electronegative metal cation in the salt. Metals that are more electronegative than uranium are not oxidized and remain in the anode basket. These metals are the transition metal fission products. Metals that are more electropositive than uranium are oxidized and accumulate in the salt as metal chlorides. These metals are the alkali, alkaline earth, and lanthanide fission products, and the transuranics. Metals that accumulate in the salt do so at the expense of the uranium chloride concentration, which has to be maintained by adding more uranium trichloride. Therefore, the electrorefiner salt chemistry keeps changing as spent fuel is processed.

Exchange reaction between UCl_3 and bond-sodium in the fuel:

$$UCl_3 + Na^0 = 3\ NaCl + U^0$$

Exchange reaction between UCl_3 and plutonium in the fuel:

$$UCl_3 + Pu^0 = PuCl_3 + U^0$$

5.2. Process Chemistry of Voloxidation

Common LWR oxide fuel is fabricated by loading high-density uranium oxide fuel pellets into cladding tubes comprised of zirconium alloys. The fuel pellets slide into the tubes. During irradiation in the reactor, mechanical and chemical bonds form between the oxide fuel and the cladding, making it difficult to separate the spent fuel from the cladding. Voloxidation is one method of improving the separation of fuel from cladding. During voloxidation, the oxide fuel is exposed to a high-temperature oxygen-enriched atmosphere to convert the uranium (IV) oxide (UO_2) into uranium (V, V, VI) oxide (U_3O_8) according to the following reaction:

$$3\,UO_2 + O_2(g) = U_3O_8.$$

This reaction results in a volume increase and decrepitation (i.e., breaking into smaller pieces) of the high-density UO_2 fuel pellets into high-surface-area U_3O_8 powder. This effect disengages the fuel from the cladding, thereby increasing the ability to separate the fuel from the cladding. In some flowsheets, the U_3O_8 powder is pelletized and loaded into the cathode baskets for the oxide reduction cell. In other flowsheets, the U_3O_8 powder is directly loaded into the cathode baskets.

Voloxidation may be used in both aqueous and pyrochemical reprocessing flowsheets for the purpose of separating the fuel from the cladding, converting the high-density UO_2 fuel pellets into U_3O_8 powder, or both.

5.3. Process Chemistry of Oxide Reduction

When pyrochemical reprocessing is applied to oxide fuel, the oxides are converted to metals in the oxide reduction cell as shown in Figure 9 [11, 12]. The reduced metals are advanced to the uranium electrorefining cell that was described earlier. The cathode is a steel basket that holds the oxide fuel, and the anode is a metal or graphite electrode that liberates the dissolved oxygen in the salt as oxygen gas ($O_2(g)$). The cathode basket and anode are submerged in a salt comprised of lithium chloride (LiCl) with a nominal bulk concentration of lithium oxide (1 wt% Li_2O). The salt is maintained at 650°C. An external DC power supply is used to pass electrical current between the cathode and anode.

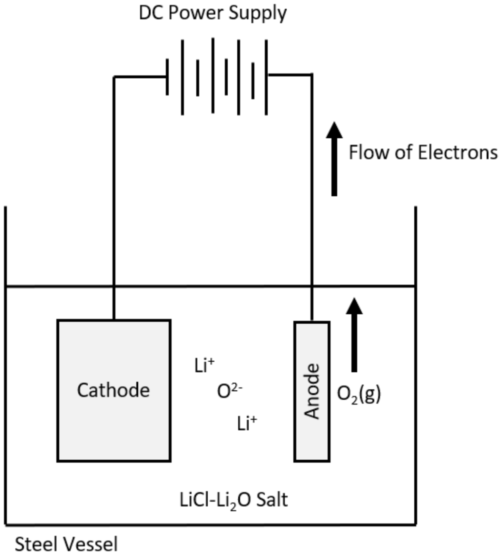

Figure 9. Simple schematic of an oxide reduction cell.

The molten salt is an ionic liquid that serves as the electrolyte to facilitate the chemical reactions. At the cathode, uranium oxide (either UO_2 or U_3O_8) is reduced to uranium metal (U^0) by liberating the chemically bound oxygen into the salt as an oxygen anion (O^{2-}). The oxygen anions migrate through the salt from the cathode to the anode. At the anode, the oxygen anion is oxidized to oxygen gas (O_2(g)).

Reduction at the cathode:

$$UO_2 + 4\,e^- = U^0 + 2\,O^{2-}$$

$$U_3O_8 + 16\,e^- = 3\,U^0 + 8\,O^{2-}$$

Oxidation at the anode:

$$2\,O^{2-} = O_2(g) + 4\,e^-$$

Electrical charge flows through the external circuit as the migration of electrons and through the salt as the migration of anions.

The spent fuel contains a large array of elements created by fission and transmutation processes. In the uranium electrorefining cell, uranium was the most electronegative cation in the salt. In the oxide reduction cell, lithium is the most electronegative cation in the salt. Only those few metals that are more electropositive than lithium can be oxidized and accumulate in the salt as metal chlorides. Two fission product metals that meet this criterion are strontium and cesium.

Exchange reaction between LiCl and cesium in the fuel:

$$LiCl + Cs^0 = CsCl + Li^0$$

It is then possible for the resulting lithium metal to reduce uranium oxide in the cathode basket:

$$UO_2 + 4\,Li^0 = U^0 + 2\,Li_2O,$$

$$U_3O_8 + 16\,Li^0 = 3\,U^0 + 8\,Li_2O.$$

5.4. Process Chemistry of Salt Distillation

Salt distillation is applied to separate salts from metals. The recovered salts are returned to their related electrochemical cells. Vacuum retort furnaces are commonly used for salt distillation. The salt-laden materials are loaded into a process crucible inside the furnace. The furnace is sealed and evacuated. Heat is applied to vaporize the salt at temperatures between 800 and 1300°C. The salt vapor collects at a cooler part of the furnace and pours into a receiver crucible. The metal products remain in the process crucible.

Fractional distillation is a separations process based on difference in vapor pressures. Substances with a higher vapor pressure and separated from substances with a lower vapor pressure as the temperature is increased. Chloride salts will undergo fractional distillation because different metal chlorides have different vapor pressures. A consequence of fractional distillation is that the composition of the salt remaining in the process crucible changes as salt is discharged from the crucible. This effect can influence the chemical separations between the salt and the metal in the process crucible, which tends to increase the partitioning of impurities into the metal.

5.5. Process Chemistry of the Liquid Cadmium Cathode

The liquid cadmium cathode (LCC) technology is used to recover transuranics from the uranium electrorefiner salt as an alloy with uranium [13, 14]. The principle of operation is based on the different chemical behaviors of uranium and plutonium (and minor actinides) in cadmium at 500°C. Cadmium that is saturated with uranium is in equilibrium with uranium metal, and the chemical activity of uranium in uranium metal is one. On the other hand, cadmium that is saturated with plutonium is in equilibrium with a plutonium/cadmium ($PuCd_2$) intermetallic, and the chemical activity of plutonium in this intermetallic is much less than one. This behavior of chemical activities allows uranium and plutonium to be reduced into the LCC at the same reduction potential. A schematic diagram of the LCC arrangement is shown in

Figure 10. Typically, the anode is loaded with spent metal fuel during LCC operations. Therefore, as uranium is oxidized into the salt at the anode, both uranium and plutonium (and minor actinides) are reduced into the cadmium pool at the cathode.

Oxidation at the anode:

$$U^0 = U^{3+} + 3\ e^-$$

Reduction at the LCC:

$$U^{3+} + 3\ e^- = U^0$$

$$TRU^{3+} + 3\ e^- = TRU^0$$

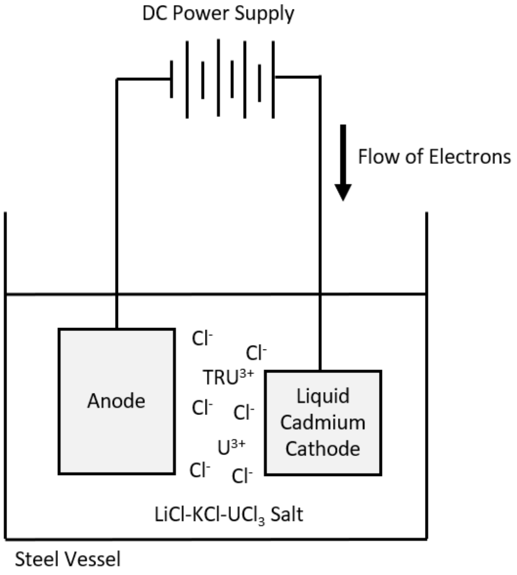

Figure 10. Simple schematic of an LCC in a uranium electrorefining cell.

5.6. IFR Flowsheet Description

The flowsheet envisioned under the IFR Program to demonstrate reprocessing of EBR-II driver fuel (i.e., HEU metallic alloy fuel) is shown in Figure 11. EBR-II was a pool-type SFR. Under this scenario, the hexagonal subassemblies containing driver fuel were removed from the sodium pool and loaded into a shieled transport cask. The cask was taken to the sodium wash station where the sodium was removed from the external surfaces of the subassembly. The cask was then taken to the air-atmosphere hot cell where the subassembly was transferred into the hot cell. Here the subassembly was dismantled to remove the fuel elements, which were loaded into carrousels for further handling inside the hot cell. The stainless steel hardware from the subassembly was waste.[d] The carrousels were moved to the element chopper in the argon atmosphere hot cell, where the spacer wires were removed in preparation of chopping. The element chopper cut the fuel elements into small pieces along the length of the fuel region. The chopped fuel pieces, now called *segments*, were loaded into the anode basket. The wires and plenums from the fuel elements were waste.[e] The anode basket was loaded into the electrorefiner.

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[d] Stainless-steel hardware from disassembled subassemblies is remote-handled low-level waste (RH-LLW).

[e] The plenum section of the fuel element is a void space that is provided to accommodate the accumulation of fission product gases without over-pressuring the fuel element. Untreated plenum sections are remote-handled high-level waste (RH-HLW). However, if treated to remove the residual bond-sodium, they may become RH-LLW.

Once electrorefining was completed, the anode residue, now called *hulls*, was advanced to a furnace to distill and collect the salt. Once electrorefining was completed, what remains in the anode basket is called anode residue, and the segments are now called hulls. Therefore, anode residue is a mixture of salt, hulls, and unoxidized fuel and fission products. The anode residue was advanced to a furnace to distill and collect the salt, which was returned to the electrorefiner. The remaining anode residue was waste. The electrorefined uranium was advanced to a furnace to distill and collect the salt and consolidate the uranium dendrites into an ingot. Some zirconium may be collected along with the electrorefined uranium. The uranium ingot was advanced to a fuel pin casting furnace where additional HEU and/or zirconium were added as needed to make the fuel alloy composition. The fuel pins were advanced to fuel fabrication to make new fuel elements, which were used to make new subassemblies for return to the reactor.

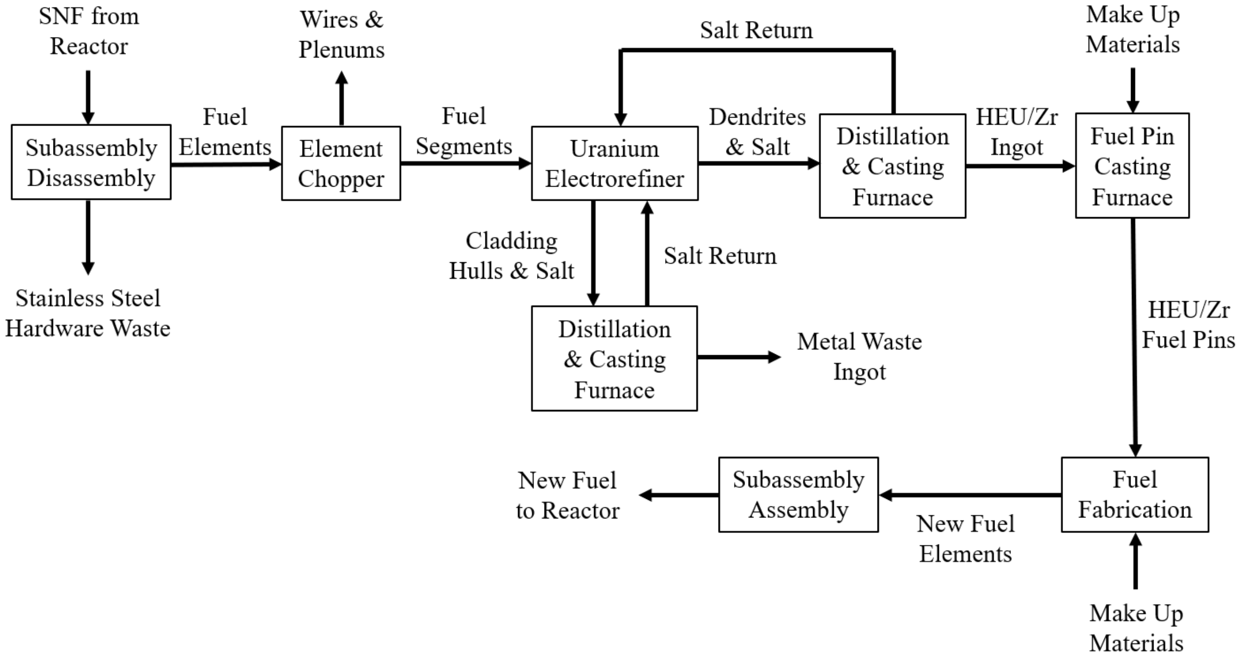

Figure 11. Pyrochemical flowsheet representing the IFR reprocessing concept.

Plutonium (and minor actinides) accumulates in the electrorefiner salt as metal chlorides. The LCC technology (not shown in Figure 11) was used to recover transuranics from the salt as an alloy with uranium. The alloy collected by the LCC is advanced to a furnace to distill and collect the cadmium along with the salt and consolidate the remaining TRU-containing alloy into an ingot. The ingot was then used for the fabrication of ternary alloy (U/Pu/Zr) fuel. However, as mentioned earlier, this flowsheet in its entirety was never demonstrated because EBR-II was shut down before the demonstration could begin.

5.7. SFT Flowsheet Description

The flowsheet presently used for the SFT Program to disposition EBR-II driver fuel and blanket materials is shown in Figure 12. When EBR-II was shut down in 1994 and the IFR Program was terminated, the flowsheet shown in Figure 11 was converted to the flowsheet shown in Figure 12. The modifications included the installation of a second chopper and electrorefiner dedicated to blanket materials and the removal of the fuel fabrication equipment that was intended to be used to make new fuel elements and subassemblies. Another modification was the practice of HEU down-blending to LEU by the addition of DU in the electrorefiner, distillation furnace, and casting furnace. The three locations of DU additions are identified on the flowsheet. The target enrichment level of this process is 19.75 wt% U^{235} in the final LEU ingots and must be less than 20 wt%. The final LEU product is cast into small or large reguli (i.e., metallic) ingots that weigh 3 or 7 kg, respectively.

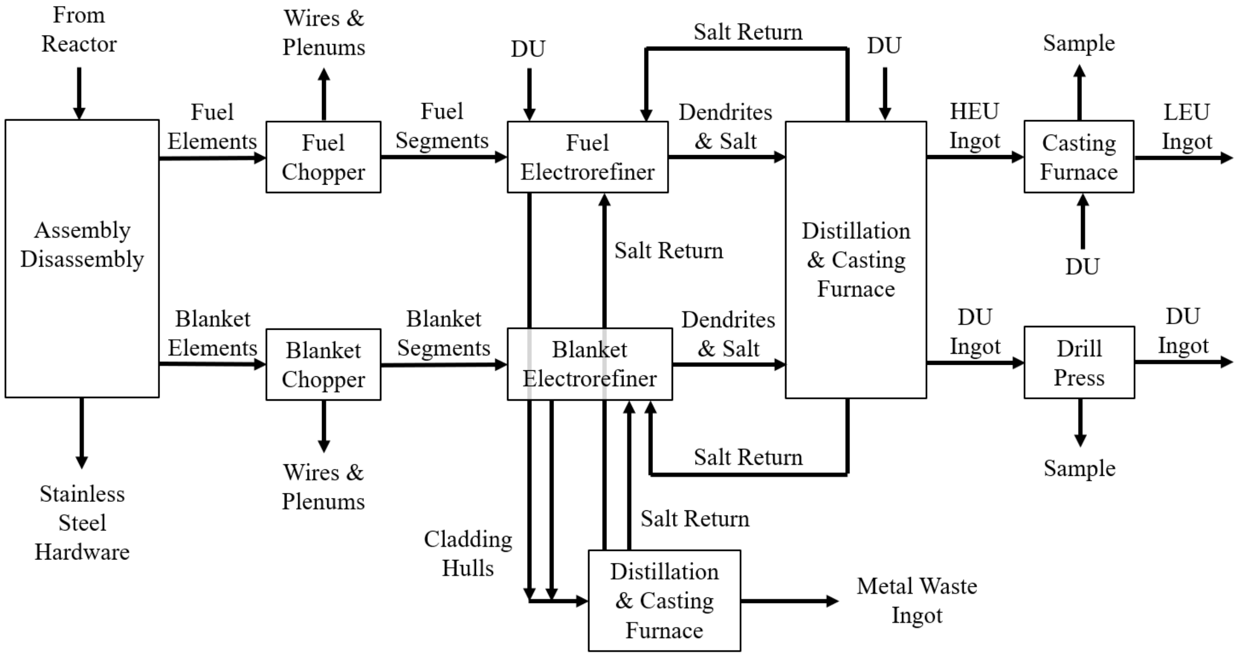

Figure 12. Pyrochemical flowsheet representing the SFT process.

5.8. JFCS Flowsheet Description

The flowsheet presently being studied for the JFCS Program to recover plutonium (and minor actinides) as a uranium/plutonium alloy to make metallic fast reactor fuel is shown in Figure 13. This process is designed for spent uranium oxide fuel clad in a zirconium alloy. Fuel decladding is the operation of splitting the fuel cladding to remove and collect the uranium oxide fuel as a crushed powder. Voloxidation is an optional process. As described earlier, voloxidation is the high-temperature processing of the fuel in an oxygen-enriched atmosphere to convert the uranium (IV) oxide (UO_2) into uranium (V, V, VI) oxide (U_3O_8). Oxide reduction is the process of electrochemically reducing the uranium oxide to uranium metal, which liberates the oxygen from the salt as oxygen gas ($O_2(g)$). The metal product from the oxide reduction cell is advanced to salt distillation that does not include metal consolidation. The distilled salt is returned to the oxide reduction cell. The metal is advanced to the electrorefining cell. The purpose of this intermediate salt distillation step is to minimize the carryover of lithium oxide (Li_2O) in the oxide reduction salt to the electrorefiner salt, where it can oxidize uranium.

Once electrorefining is completed, the anode residue is advanced to a furnace to distill and collect the salt. The salt is returned to the electrorefiner, and the anode residue is waste. The electrorefined uranium is advanced to a furnace to distill and collect the salt and consolidate the uranium dendrites into an ingot. Because uranium in spent LWR fuel is only slightly more enriched than natural uranium, the electrorefined uranium that is recovered by this process may or may not have value.

Plutonium (and minor actinides) accumulates in the electrorefiner salt as metal chlorides. The LCC technology is used to recover transuranics from the salt as an alloy with uranium. The objective of LCC operations is to recover an alloy that is approximately 50 wt% uranium and 50 wt% transuranics. The alloy collected by the LCC is advanced to a furnace to distill and collect the cadmium along with the salt and consolidate the TRU-containing alloy into an ingot. The ingot is then used for the fabrication of ternary alloy (U/Pu/Zr) fuel.

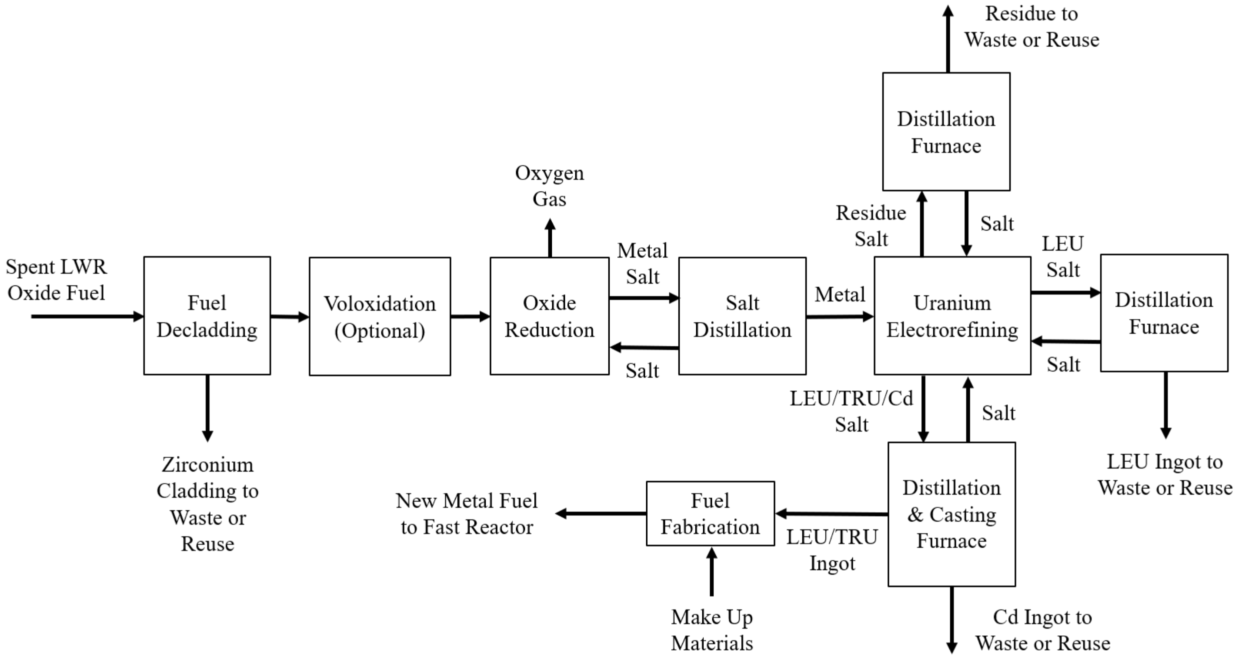

Figure 13. Pyrochemical flowsheet representing the JFCS process.

6. AQUEOUS REPROCESSING OFF-GAS POINT SOURCES

Gaseous emissions from the aqueous reprocessing of nuclear fuel can be categorized according to the specific unit operation from which they arise. Figure 14 shows a schematic diagram of how these emissions might be categorized in the context of a standard aqueous flowsheet [15].

Upon receipt, spent UO_x-type nuclear fuel is debundled and transferred to a mechanical shearing operation. As the mechanical shear breaches the cladding, any free gases contained within the fuel element and not physically bound with the fuel pellets can be released into the hot cell and associated hot cell ventilation system. The size of the mechanical equipment used for the shearing operation often dictates that shearing is conducted in an open, dedicated hot cell. Therefore, all gases are released into the open cell, and the volatile fission products become highly diluted in the cell ventilation stream, also known as the cell off-gas (COG) stream, which is labeled A in Figure 14.

Following shearing, fuel and cladding pieces are transferred to the dissolver where the fuel is dissolved out of the cladding hulls by nitric acid. Off-gas arising from dissolution is not vented into the cell but is directly transferred for treatment as a separate dissolver off-gas (DOG) stream, which is labeled B in Figure 14 [16]. Gases that are released from the dissolver into the open cell become highly diluted in the COG stream, which is labeled C in Figure 14.

Uranium-bearing solution from the dissolver is then transferred to the separation processes. Although the separation processes are complex and may represent many specific engineering operations (e.g., primary separations, secondary separations, solvent recycle, acid recycle), they are typically treated as a single off-gas stream that arises from the combined venting of all the separation vessels. This combined stream has often been referred to as the vessel off-gas (VOG) stream, which is labeled D in Figure 14.

Finally, there are a variety of waste operations associated with spent fuel reprocessing, including vitrification, calcination, liquid waste treatment, and solids compaction. These streams can be managed in different ways, but there is some limited source-term information specific to the vitrification step conducted within the glass melter. Off-gas streams arising from waste processing have been referred to as waste off-gas (WOG) stream, melter off-gas (MOG) stream, and other similar terms. In Figure 14, the off-gas from solvent treatment is labeled E, and the off-gas from liquid waste solidification is labeled F.

Eventually the treated off-gas streams are combined before being discharged to the atmosphere through a stack. This combined stream is labeled G in Figure 14.

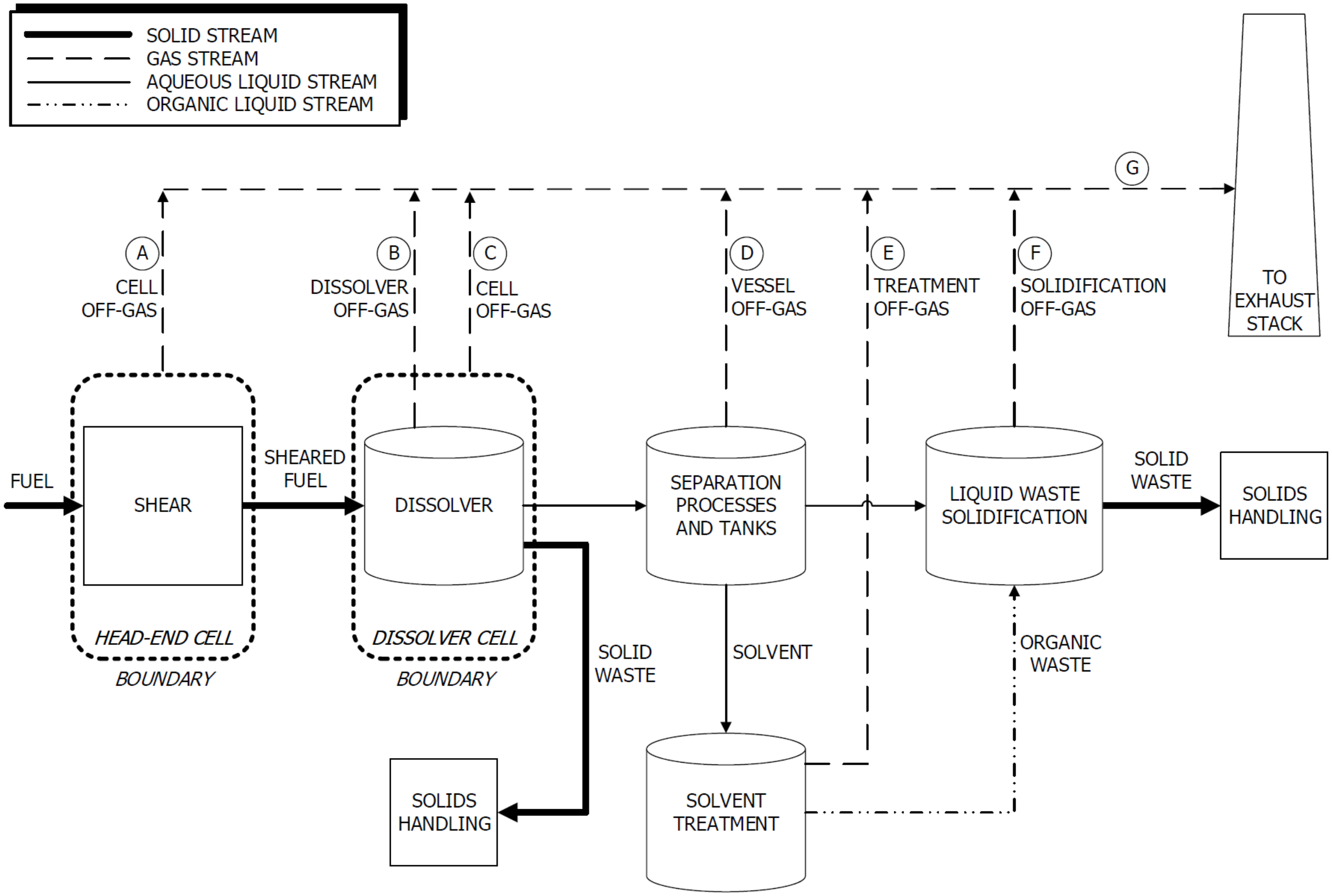

Figure 14. Schematic diagram showing aqueous reprocessing off-gas streams.

6.1. Voloxidation

Figure 14 describes the most common implementation of an aqueous reprocessing flowsheet and its associated gaseous emissions, but there is one alternative implementation that merits discussion due to its significant impact on off-gas treatment. Figure 15 shows an aqueous flowsheet that incorporates a voloxidation step prior to fuel dissolution. As described earlier in the context of UO_x fuel, voloxidation refers to exposure of the spent fuel pellets to an oxidizing gas stream (most commonly O_2), thus converting solid UO_2 to U_3O_8 powder according to $3\ UO_2(s) + O_2(g) = U_3O_8(s)$. The addition of a voloxidation step is expected to provide benefits to an aqueous flowsheet in at least two aspects. First, comminution (i.e., size reduction) of the ceramic fuel pellets into a fine powder increases dissolution rate (decreases dissolver cycle time). Second, voloxidation releases key gaseous elements (especially tritium and the noble gases) in a controlled manner, allowing them to be captured early in the process before they distribute into multiple plant ventilation streams.

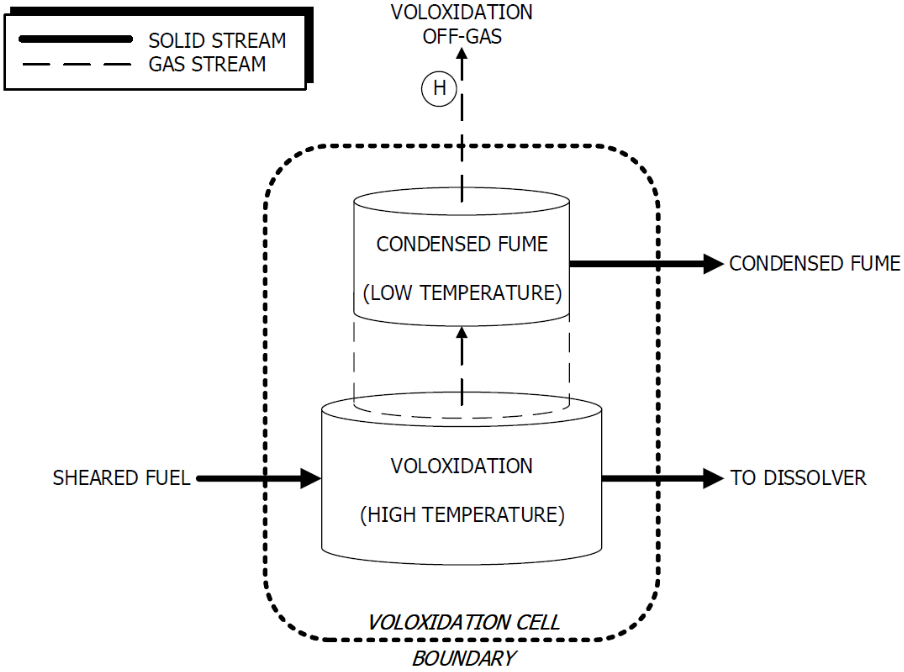

Figure 15. Schematic diagram showing voloxidation off-gas streams.

In Figure 15, *Condensed Fume* is material that entered the off-gas stream due to its high vapor pressure at the operating temperature of the voloxidation process. As the off-gas cools, the vapor condenses into a fume that will likely be captured by filters. The *Condensed Fume* represents a waste stream containing semivolatile fission products that must be managed.

In a typical aqueous flowsheet without voloxidation, tritium transfers from the SNF to the aqueous phase as tritiated water or other aqueous species during dissolution. From there, some fraction distributes to the off-gas stream as vapor, while the remainder transfers through to separations. In an operating plant that utilizes acid recycle, tritium will distribute to nearly all aqueous streams and most off-gas streams, adding complexity to treatment [17]. The addition of the voloxidation step can mitigate this concern substantially by releasing tritium in a single step early in the recycle process. Other permutations of voloxidation exist, with the most common being the use of NO_2 gas as an oxidant. This NO_2-based voloxidation releases both iodine and tritium into the gas phase, allowing both to have limited distribution through the plant. The technology associated with NO_2 voloxidation is not yet developed to the point of commercial implementation.

6.2. Tritium Release Pathways

The primary release pathway for gaseous tritium-bearing species in a standard reprocessing flowsheet is through the dissolution process. As tritium is released from the fuel, it transfers into the concentrated nitric acid solution and exists as tritiated water or tritiated nitric acid ($^3HNO_3$). As dissolution is conducted at an elevated temperature, the vapor pressure of both water and nitric acid will allow transfer of the tritium-bearing species to the gas phase and subsequent transfer to the DOG as the dissolver vessel is vented. Typically, a condenser or series of condensers is placed very early in the DOG treatment system to condense these vapors back into the dissolution vessel. The operating temperature of these condensers will determine the specific fraction of tritium that remains in the gas phase vs. returns to the dissolver vessel [18]. After the condensers, tritiated water constitutes the primary tritium-bearing species present in the off-gas.

As mentioned earlier, voloxidation can be used to release tritium prior to dissolution. In this case, tritium is released nearly completely from the fuel as tritiated water and can be subsequently captured using standard moisture removal techniques such as molecular sieves.

6.3. Semivolatiles Release Pathways

Semivolatiles is a term that generally refers to elements that do not have significant vapor pressure in their elemental state at ambient temperature. As such, semivolatile release pathways are somewhat limited to high-temperature operations, although in some cases the semivolatile element is able to form a chemical species with high volatility at relatively low temperatures. One of the most interesting isotopes is Ru-106 [19]. Ru is expected to have releases associated with voloxidation, vitrification, and dissolution, but estimates for specific release values have wide variance and merit additional measurements [20]. Although dissolution is the lowest temperature process of the three just listed, it is included because evidence shows the accumulation of ruthenium in DOG treatment systems. A mechanism for formation and release of ruthenium from the dissolver was identified [21]. In high-nitrate media, Ru volatilization occurs through formation of Ru-nitrate species that can be subsequently oxidized to volatile RuO_4.

6.4. Noble Gas Release Pathways

Noble gases (most notably, Kr and Xe) are released early in the recycling flowsheet. Some small fraction is released from the fuel pins during shearing due to noble gas migration out of fuel pellets during irradiation. The balance of the noble gases is completely released during dissolution (or voloxidation) of pellets when the fuel meat itself is broken down.

6.5. Iodine Release Pathways

Iodine release pathways are the most complex of the elements discussed here. Iodine is released by most processes in the flowsheet and can present as different species. Only a small fraction of iodine is released during shearing, with most of the iodine present in the fuel meat following irradiation.

The most significant source of iodine release is the dissolver. The dissolver is expected to release a significant amount of iodine into the vapor phase, with the remainder present in the dissolver solution or as undissolved solids within the dissolution vessel. Studies with simulated fuel solutions indicate release fractions ranging from 93–99%, but a study using spent fuel indicated a lower volatile fraction, with > 10% of iodine remaining in either the dissolver solution or as undissolved solids [22, 23]. The amount of iodine released from the dissolver can be increased through certain sparging techniques. The iodine released from the dissolver is primarily I_2, although other inorganic species like hypoiodous acid (HOI) may be present.

Iodine is further released into the VOG stream during separations as the dissolver solution is processed; 1–5% of iodine is predicted to be released during separations and may have complex speciation, with elemental iodine (I_2) and organic iodides present in the off-gas. These organic iodides may include a range of species such as methyl iodide (CH_3I), butyl iodide (C_4H_9I), and dodecyl iodide ($C_{12}H_{25}I$) [15]. Organic iodides have been historically considered more challenging to treat than elemental iodine, and the United States has performed a significant amount of R&D on potential abatement mechanisms.

Any iodine remaining in solution can volatilize during vitrification or calcination as elemental iodine, mixed halides, or in a complex with nitric acid/nitrate species. Release fractions and speciation during waste treatment are not well understood.

Finally, iodine may have some release during air/O_2 voloxidation, but near complete release is expected during NO_2-based voloxidation, which would then subsequently decrease the amounts seen downstream of those operations.

7. PYROCHEMICAL REPROCESSING OFF-GAS POINT SOURCES

In its present state, pyrochemical reprocessing is a manually operated batch process where materials are passed between unit operations in a batchwise fashion by facility operators. However, as pyrochemical reprocessing technologies are scaled up to commercial capacities, advances in mechanical design are expected to introduce automation into the processes. The descriptions presented here are of unit operations that are largely independent of each other with respect to materials flow, as materials leaving a downstream unit operation may be accumulated in storage (i.e., a form of surge capacity) before being advanced to the next upstream unit operation.

Figure 16 to Figure 26 represent different unit operations that can be used in a pyrochemical reprocessing flowsheet. All the figures are similar in presentation, but each represents a different set of circumstances with respect to the management of dust, fume, and off-gas. Dust is fine particulate. Condensed fume is also fine particulate but forms as a high-temperature gas stream (containing condensable vapor) cools. Off-gas is anything that is a gas at high temperatures and remains a gas at ambient pressures and temperatures.

7.1. LWR Oxide Fuel

Mechanical decladding is shown in Figure 16, where oxide fuel is separated from the zirconium alloy cladding. This operation releases noble gas fission products into the off-gas due to breaching the fuel cladding and creates dust due to the mechanical action of decladding. Filtration can capture the dust as the noble gas fission products report to the off-gas stream.

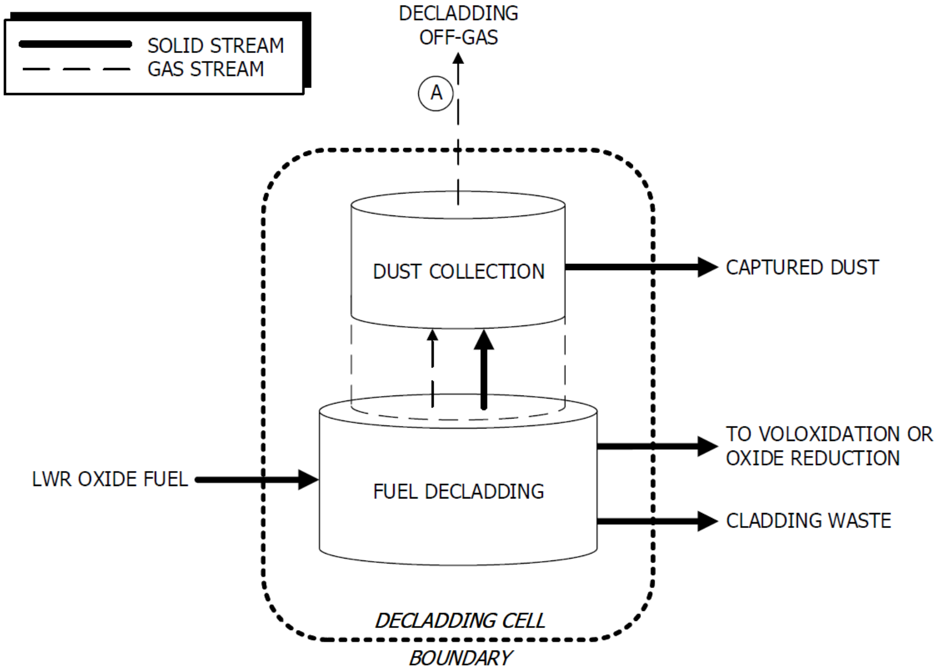

Figure 16. Schematic diagram of mechanical decladding off-gas.

Voloxidation is shown in Figure 17, which, as discussed earlier, converts UO_2 fuel pellets to U_3O_8 powder in a high-temperature operation involving oxygen gas. This operation releases noble gas fission products due to the high temperature and decrepitation of the UO_2 fuel as it converts to U_3O_8. This operation also releases semivolatile fission products due to the vapor pressure of these species at high temperature. As the off-gas cools, the semivolatile fission products will condense into a fume. Filtration can capture the dust and fume as the volatile fission products report to the off-gas stream.

A possible candidate for the process gas delivered to the furnace is a mixture of purified argon and oxygen. In this case, only a portion of the oxygen will be consumed in the process, and the off-gas stream will contain the argon and the excess oxygen provided to the furnace.

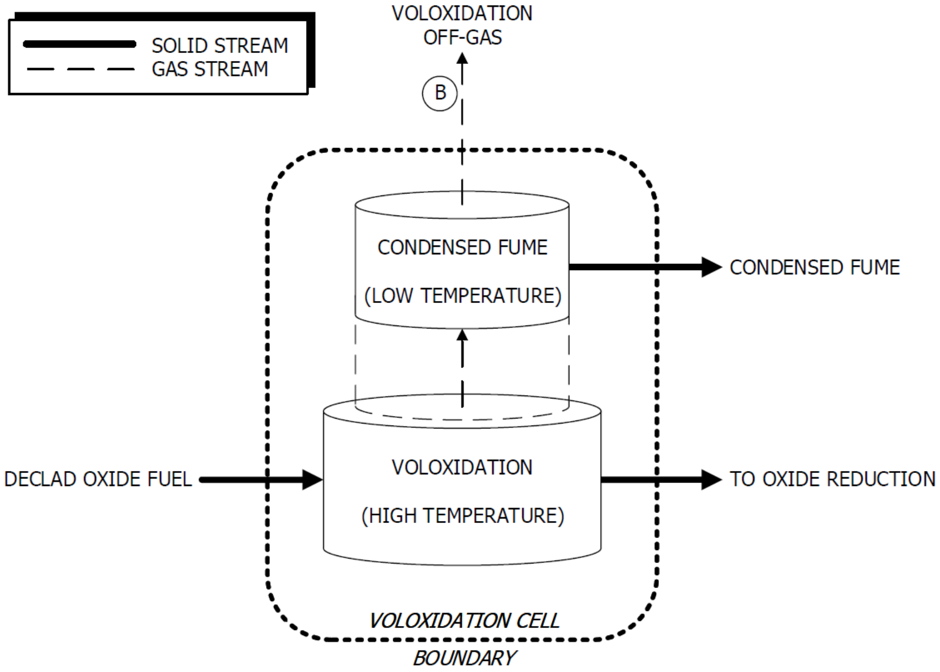

Figure 17. Schematic diagram of voloxidation off-gas.

Cathode basket loading is shown in Figure 18. This operation is representative of any materials handling operation involving fine particles. Such operations include collecting the declad fuel, sampling and sizing the declad fuel, loading the declad fuel into the voloxidation furnace, unloading the voloxidation furnace, loading the cathode basket, cleaning the cathode basket, and managing the collection of dust and fume from the various unit operations. Some flowsheets propose pelletizing the oxide fuel from the voloxidation furnace prior to loading the cathode baskets. All these operations will likely be challenging with regards to dust containment and collection. The release of volatile fission products into the off-gas is expected to be minimal.

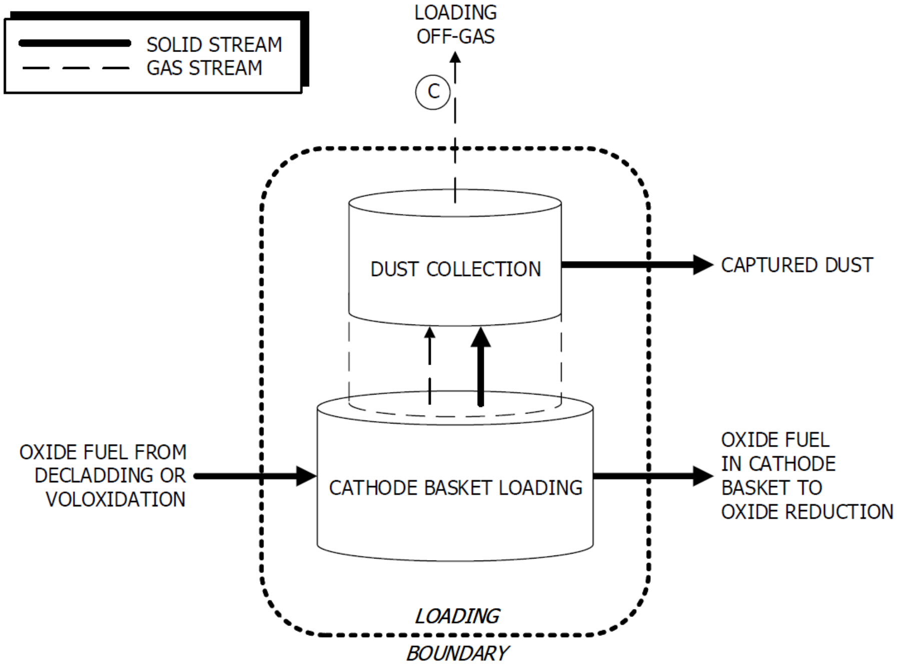

Figure 18. Schematic diagram of cathode basket loading off-gas.

Oxide reduction is shown in Figure 19, which, as discussed earlier, converts uranium oxide to uranium metal by means of electrochemical reactions in a high-temperature molten salt. This operation releases the remaining noble gas fission products from the oxide fuel. The salt is composed of LiCl with a small concentration of Li_2O. Select fission products will accumulate in the salt as metal halides. These elements are predominantly rubidium and cesium (alkali), strontium and barium (alkali earth), tellurium (metalloid), iodine (halide), and europium (lanthanide). Various mechanisms have been proposed for how these fission products accumulate in the oxide reduction salt [24].

By virtue of the high temperature, everything in the salt has a vapor pressure above the salt. As these vapors follow the off-gas stream, different salts will condense at different temperatures in a process called fractional distillation. Some salts will condense in some locations as liquids and drain back into the vessel. Other salts will condense as solids and adhere to the cooler parts of the process equipment. The main point of Figure 19 is to show that both off-gas and condensed fumes will have to be managed.

The oxide reduction process generates oxygen gas at the anode. This oxygen gas is the fate of the oxygen associated with the incoming uranium oxide feed to the oxide reduction cell. As the uranium oxide is reduced to uranium metal, the oxygen is liberated as oxygen gas at the anode. It is possible that the headspace above the salt will be continuously purged with a stream of purified argon for the purpose of directing the flow of oxygen away from the salt and to provide a positive pressure above the salt relative to the surrounding atmosphere.

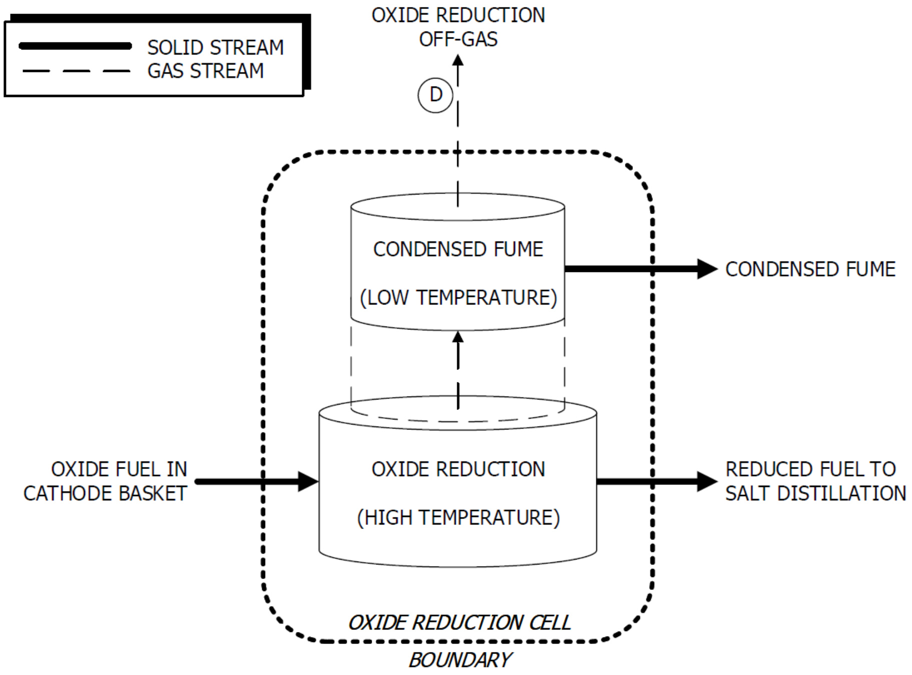

Figure 19. Schematic diagram of oxide reduction off-gas.

To prevent the carryover of salt from the oxide reduction cell to the uranium electrorefining cell, the reduced metal from the oxide reduction cell is processed in a salt distillation furnace as shown in Figure 20. In this process, the salt vapor is captured and condensed so that it can be returned to the oxide reduction cell. The load of volatile fission products reporting to the off-gas is expected to be minimal.

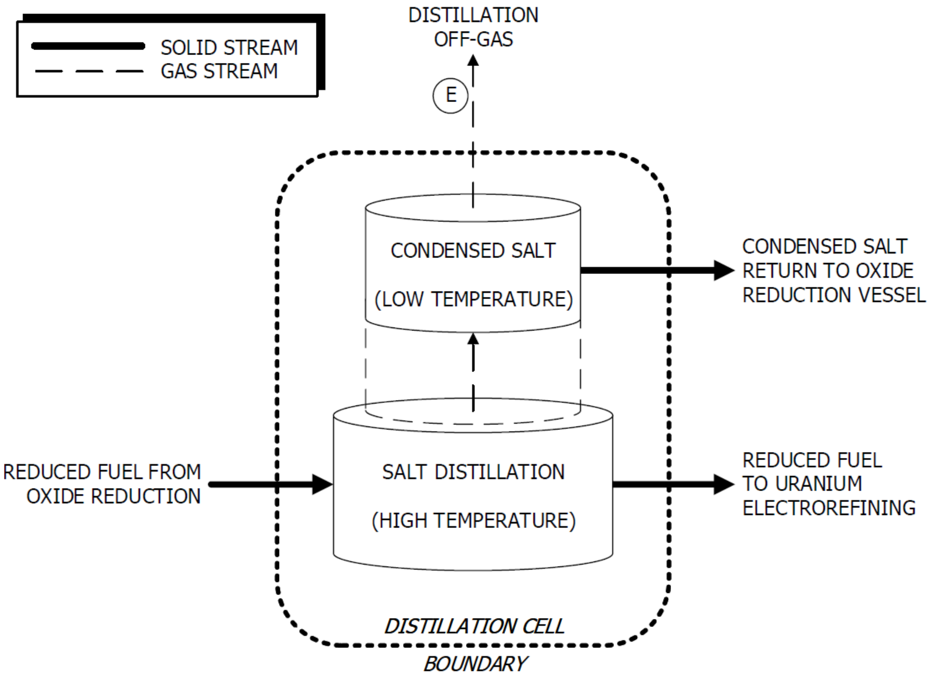

Figure 20. Schematic diagram of salt distillation off-gas.

7.2. SFR Metallic Fuels

The unit operations described in Figure 16 to Figure 20 are associated with LWR oxide fuels. The processing of SFR metal fuels begins with washing the sodium from the surfaces of the fuel assembly hardware as shown in Figure 21. Fuel assemblies removed from a pool-type SFR will be coated in sodium. Residual sodium coats the surfaces of the fuel ducts, fuel elements, and other associated hardware. Sodium metal is highly reactive with water. Therefore, sodium wash systems are engineered to mitigate the thermal, explosive, and radiological hazards. The residual sodium is removed by washing the hardware with denatured ethyl alcohol, wet nitrogen gas, steam, or water [25, 26, 27]. This sodium is subject to fission product contamination by mechanisms such as tritium capture and fuel cladding failures within the sodium pool [28]. Therefore, the wash solutions must be managed as radiological solutions. The off-gas stream will contain hydrogen gas and possibly tritium.

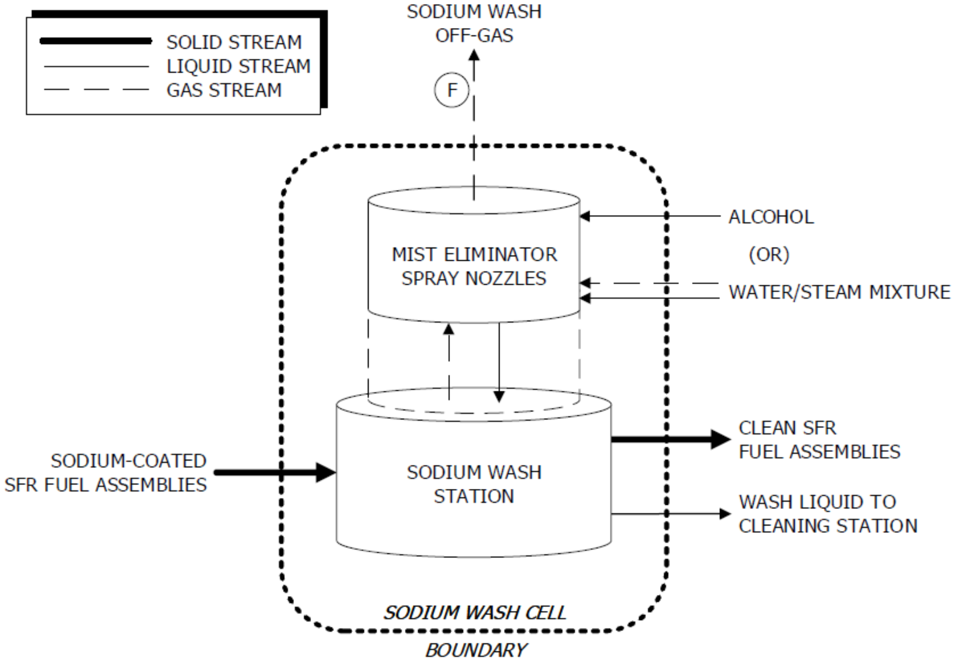

Figure 21. Schematic diagram of sodium washing off-gas.

For pyrochemical reprocessing, it is generally anticipated that LWR oxide fuel will undergo fuel decladding, and that SFR metal fuel will undergo fuel chopping as shown in Figure 22. This operation releases noble gas fission products due to breaching the fuel cladding and creates particles (debris) due to the mechanical shearing operations. Barrier and filtration can capture the particles as the noble gas fission products report to the off-gas stream. Chopping sodium-bonded metal fuels will create debris containing sodium splatter and small pieces of fractured fuel and cladding. The remaining plenum sections will contain some fraction of the bond-sodium and the fission products that dissolve into the bond-sodium. Management of plenum sections can result in another unit operation to neutralize the entrained sodium metal.

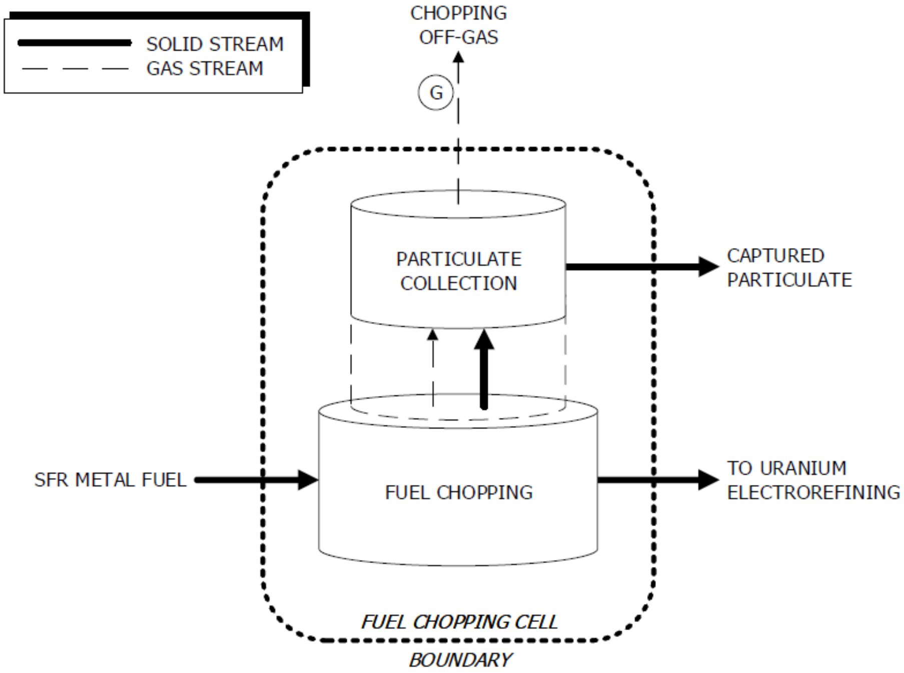

Figure 22. Schematic diagram of fuel chopping off-gas.

Many metallic fuels contain bond-sodium within the fuel element, which can collect fission products such as cesium and iodine. One disposition path for the bond-sodium is to react it with the salt in the uranium electrorefiner. The bond-sodium will accumulate in the electrorefiner salt as sodium chloride ($NaCl$) by displacement of uranium chloride (UCl_3) from the salt. An alternative disposition path is to distill much of the bond-sodium away from the chopped fuel and plenums in a sodium distillation furnace as shown in Figure 23. Keeping bond-sodium out of the electrorefiner has the benefit of reducing salt waste. However, distillation produces a sodium distillate waste that is highly pyrophoric and must be managed by other means. The sodium distillate can be converted to less reactive forms of sodium such as sodium oxide (Na_2O), sodium chloride ($NaCl$), sodium hydroxide ($NaOH$), and sodium carbonate (Na_2CO_3).

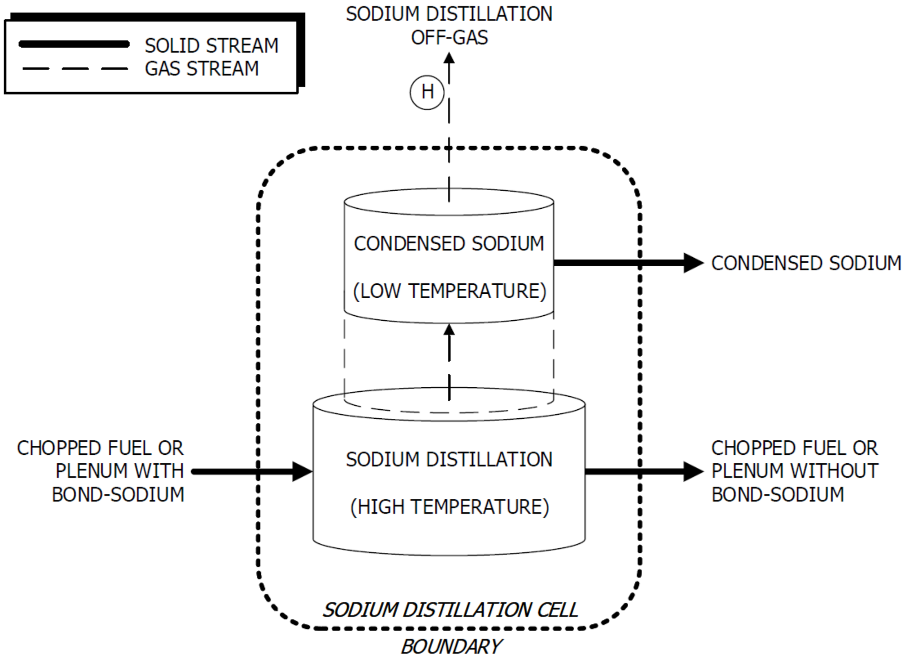

Figure 23. Schematic diagram of sodium distillation off-gas.

The uranium electrorefiner accepts uranium metal from both LWR oxide fuel processing and SFR metal fuel processing. Uranium metal from LWR oxide fuel processing was created in the oxide reduction cell and is expected to be virtually free of noble gas fission products. However, uranium metal from chopped SFR metal fuel will contain noble gas fission products. Electrorefiner salt is composed of LiCl-KCl eutectic with a nominal concentration of UCl_3. By virtue of UCl_3 in the salt, significant chemical separations occur during electrorefining. Fission products that are more electropositive than uranium accumulate in the salt. These metals include the alkali, alkaline earth, halides, lanthanides, and transuranics. Fission products that are more electronegative than uranium remain in the anode basket. These metals include the transition metals [29].

Each component in the salt will have an elevated vapor pressure as a result of the high-temperature processing. As these vapors follow the off-gas stream, different salts will condense at different temperatures in a process called fractional distillation. Some salts will condense in some locations as liquids and drain back into the vessel. Other salts will condense as solids and adhere to the cooler parts of the process equipment. The main point of Figure 24 is to show that both off-gas and condensed fumes will have to be managed.

The uranium electrorefining process does not generate gas at either the anode or the cathode. This contrasts the oxide reduction process which generates oxygen gas at the anode. It is possible that the headspace above the salt will be continuously purged with a stream of purified argon to provide a positive pressure above the salt relative to the surrounding atmosphere.

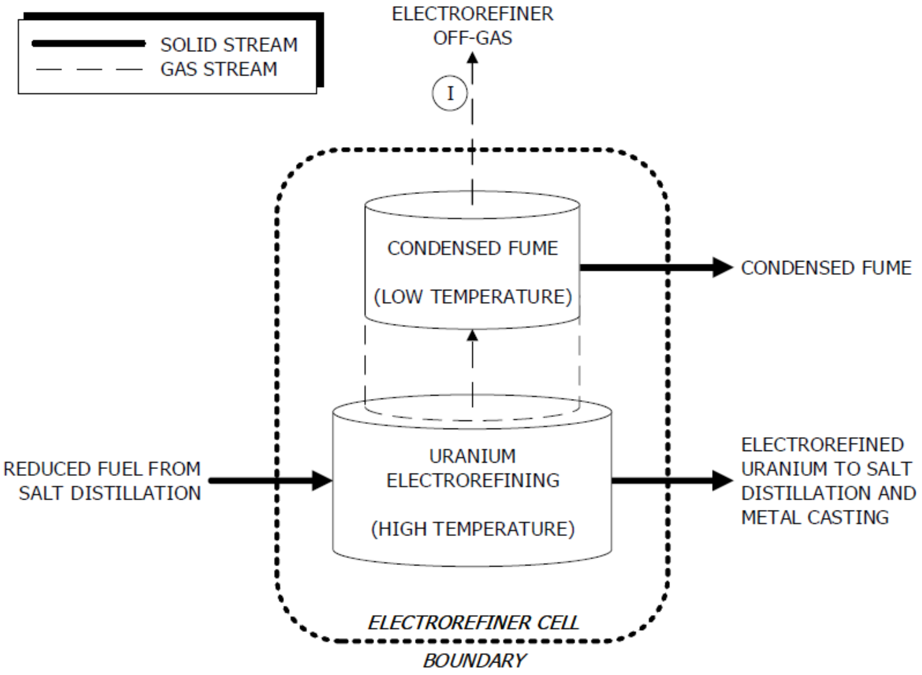

Figure 24. Schematic diagram of electrorefiner off-gas.

Electrorefined uranium has a high-surface-area dendritic morphology and includes a significant mass fraction of adhering salt. To separate the salt from the metal and consolidate the metal into an ingot, the product from the electrorefining cell is processed in a salt distillation/metal casting furnace as shown in Figure 25. In this process, the salt vapor is captured and condensed so that it can be returned to the electrorefining cell. The load of volatile fission products reporting to the off-gas is expected to be minimal.

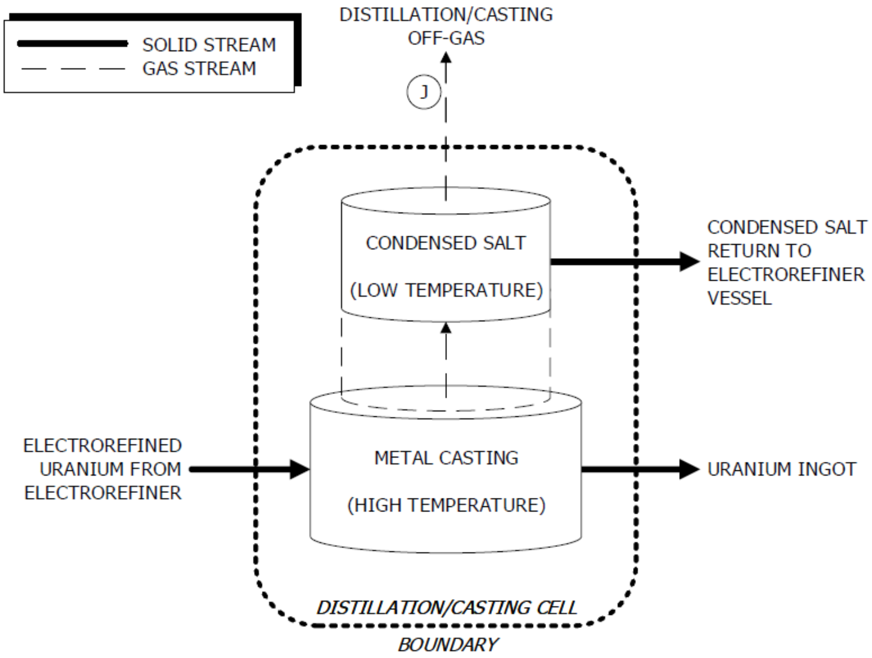

Figure 25. Schematic diagram of salt distillation/metal casting off-gas.

There may be several metal casting operations that do not involve salt as shown in Figure 26. Condensed fume and off-gas will be minimal in these operations but may still require management. There are several other operations related to materials handling, equipment maintenance, process sampling, waste disposal, etc., that can generate dust and debris that must be managed.

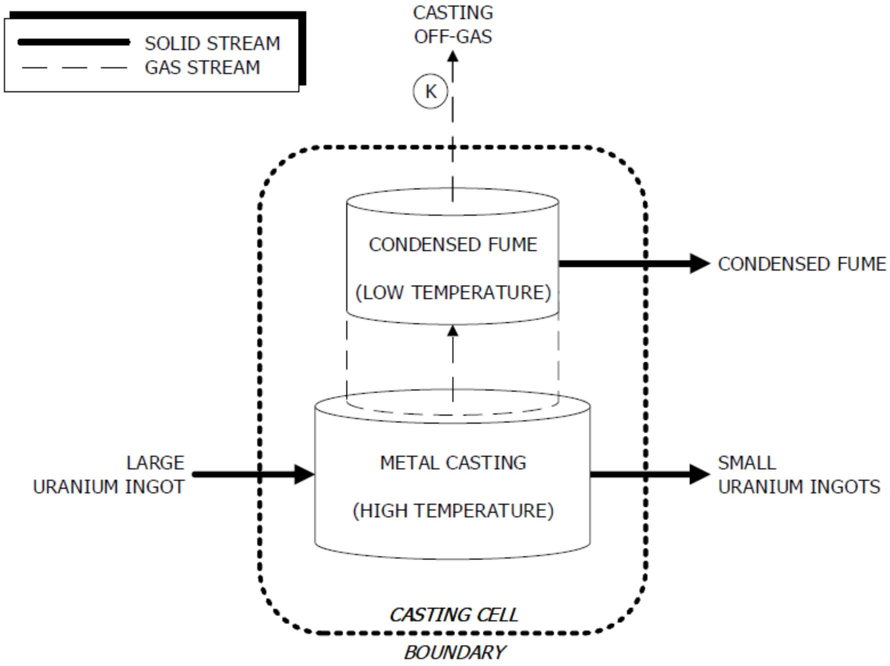

Figure 26. Schematic diagram of metal casting off-gas.

The characteristics of the unit operations shown in Figure 16 to Figure 26 are summarized in Table 2. It is assumed here that the unit operations summarized in Table 2 will be located within an argon atmosphere hot cell.[f] How all of these off-gas streams, and hot cell argon atmosphere, are managed is speculative. Some of these unit operations may provide off-gas streams that will be managed independently, while others may be combined, and others may be vented into the hot cell atmosphere.

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[f] The authors will not speculate on the design features of a future commercial scale pyrochemical reprocessing facility other than to say that there will be unit operations housed within a radiation shielded facility. The authors are experienced with the EBR-II Spent Fuel Treatment Program and the U.S./ROK Joint Fuel Cycle Study, but a future reprocessing flowsheet and facility may be something quite different.

Table 2. Summary of pyrochemical reprocessing off-gas sources.

Figure	Unit Operation	Off-Gas Stream	Major Contributions to Off-Gas Load
16	LWR Fuel Decladding	A	Breach cladding and release noble gas fission products. Dust control.
17	Voloxidation	B	High-temperature operation. Release noble gas and semivolatile fission products.
18	Cathode Basket Loading	C	Dust control.
19	Oxide Reduction	D	High-temperature operation. Release noble gas and semivolatile fission products.
20	Salt Distillation	E	High-temperature operation. Fume control.
21	SFR Sodium Washing	F	Suspect contamination and sodium reactivity control.
22	SFR Fuel Chopping	G	Breach cladding and release noble gas fission products. Particulate control.
23	Sodium Distillation	H	High-temperature operation. Sodium metal distillate.
24	Uranium Electrorefining	I	High-temperature operation. Release noble gas and semivolatile fission products.
25	Salt Distillation and Metal Casting	J	High-temperature operation. Fume control.
26	Metal Casting	K	High-temperature operation. Fume control.

8. OFF-GAS CAPTURE TECHNOLOGIES

This section provides a general introduction to various off-gas capture technologies; however, the information in the section is not intended to identify the technologies that a specific plant design should implement.

8.1. Cryogenic Distillation

Cryogenic distillation has historically been utilized to remove noble gases (specifically Kr and Xe decay products) from off-gas streams. As the name is meant to imply, noble gases have very low chemical reactivity and consequently are not removed by chemical interactions like those underpinning more common capture technologies such as adsorption or wet scrubbing. Typical implementation in nuclear reprocessing facilities places cryogenic distillation at the end of a treatment process, when most other constituents of interest have already been removed. Although Kr and Xe are recovered from air on an industrial scale, nuclear applications are distinctive because these noble gases are present at 16:1 Xe:Kr ratio (by mass) and 10:1 Xe:Kr ratio (by volume as gas) in irradiated fuel, nearly the inverse of their concentration ratios in our atmosphere.[g]

Typical implementation of cryogenic distillation involves feeding a gas mixture upward into a column with a temperature gradient. In nuclear reprocessing applications, this gas mixture is likely to include argon, nitrogen, and oxygen, with other components like NO_x and moisture removed prior to distillation [30]. Modes of operation can vary, but in some cases, Kr and Xe can be condensed to the column bottoms in liquid nitrogen carrier, transferred to a second column, and evaporated out selectively [31]. Each cryogenic distillation column provides a finite separation factor; if higher purification is desired, the gas or liquid effluents may be used to feed a second column to provide additional separation. Industrially, cryogenic distillation plants are typically referred to as an air separation unit (ASU), and industrial applications will describe their capabilities in terms of installed ASUs.

Cryogenic distillation in nuclear applications could be implemented in two ways. The first is to separate Kr and Xe from other gases and then condense the two, together, followed by transferring them to bulk gas storage. Kr-85 (10.76-year half-life) is the longest-lived of the Kr and Xe radioisotopes, and bulk decay storage could eliminate the need for dedicated waste disposition. Some solutions associated with this approach include pre-loading the Kr onto adsorbents prior to storage to provide an additional retention barrier beyond just the gas storage vessels. An alternate gas storage mechanism is to embed Kr into transition metal alloys via ion implantation [32].

The second way to apply cryogenic distillation is to remove Kr and Xe from the bulk gas stream and then separate Kr and Xe from each other, resulting in a Kr stream that contains radioactive isotopes and a Xe stream that contains stable isotopes. The isolated Kr can be managed as discussed above, while the Xe may be released or sold as a product. The decision to implement Kr/Xe separations requires weighing storage and disposal costs against the cost of extra separative units. Separation of Kr and Xe decreases the volume of waste by a factor of 10, reducing waste storage or disposal costs accordingly. However, cryogenic distillation is an extremely energy-intensive separation process, with the primary energy consumption being from the need to keep separations equipment at low temperatures.

The most notable safety concern associated with cryogenic distillation is related to the co-condensation of impurities (including oxygen, radiolytic ozone, acetylene, methane, and hydrogen) into the liquid phase. These species pose an explosion risk. In an aqueous reprocessing flowsheet, an additional contaminant of concern is breakthrough NO_x (originating from the fuel dissolution process), which can condense within the distillation columns and interfere with their function. These concerns are primarily mitigated through the addition of chemical scrubbing units upstream of the distillation column.

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[g] Atmospheric krypton is approximately 1.14 ppm (by volume), and xenon is approximately 87 ppb (by volume).

Typical scrubbing units include selective catalytic reduction of residual NO_x, catalytic O_2 and O_3 removal, and solid adsorbers to remove moisture and other contaminants [33].

While cryogenic distillation is widely utilized in industrial gas production, its use in the treatment of reprocessing effluents has been limited to pilot or demonstration scales, most notably at the Idaho Chemical Processing Plant and the Tokai Reprocessing Plant [34]. However, it is expected that any reprocessing facility in the United States will incorporate Kr recovery into their effluent treatment plan to facilitate compliance with regulatory requirements. Notably, fuel that has been cooled for an extended period (such as the existing LWR SNF inventory) will not contain Kr-85 at levels requiring treatment or capture [35, 36]. The need for Kr recovery is highly dependent on the age of the fuel being reprocessed.

For reasons including improved safety profile, decreased energy usage, and improved separations efficiency, alternatives to cryogenic distillation for Kr/Xe separations have been actively explored in the United States. These alternatives typically include the use of solid materials (i.e., molecular sieves, metal-organic frameworks [MOFs], zeolitic frameworks) to facilitate retention of either Kr or Xe at above-cryogenic temperatures [37, 38]. These materials are often employed with pressure and/or temperature swings to allow sorbent regeneration and noble gas collection.

8.2. Sorption

There are two types of sorption mechanisms employed in chemical separations: *absorption* and *adsorption*. The first, *ab*sorption, refers to a solute of interest dissolving into the volume of a solvent phase. In the context of this work, absorption would refer to solute gas molecules partitioning into a liquid or solid solvent. *Ad*sorption, more commonly found in off-gas separations, refers to the use of adsorbent materials to retain targeted elements or species. *Chemisorption* is used when retention occurs through chemical bonding of the adsorbent with the volatile target element or species; *physisorption* refers to retention through weak van der Waals interactions between the adsorbent and volatile species. Chemisorption is typically less reversible than physisorption.

The potential for both chemisorption and physisorption should be understood when testing or selecting adsorbent materials as reversibility can affect operating procedures (some materials may require purging to remove physisorbed volatile species before removal from the system), and reversible physisorption may bias measurements of adsorbent capacity for the volatile species. For all adsorption applications, there will be an operative temperature range, and the temperature is often controlled. For reversible adsorption applications, it is typical to perform adsorption at lower temperatures and then to raise the adsorbent temperature when desorption is desired.

Diverse types of materials can be used as adsorbents (e.g., minerals, impregnated charcoals, engineered frameworks), and adsorbents will be selected based on their affinity for the specific volatile species of interest. However, adsorbents can all be understood using a few key measures of performance [39]. The primary performance metric of interest is always the adsorbent's capacity for the volatile species in the operational temperature and pressure range. Capacity refers to the amount of material that can be adsorbed by a unit mass (or unit volume) of the adsorbent. Capacity of an adsorbent can vary widely across different adsorbent/adsorbate combinations. Adsorbent capacities are best understood by comparison to other adsorbents targeting the same adsorbate (e.g., some adsorbents may have capacities of 100 mg/g for a specific target volatile, while other adsorbents may have capacities of 1 mg/g for a different target volatile—both capacities may be acceptable in the context of their particular application). Capacity can be dependent on several operating variables such as temperature, pressure, and off-gas flow rate. Higher capacities can have special benefit in nuclear applications as they minimize the volume of used material requiring storage and disposal.

Beyond capacity, there are many other parameters that are important in evaluating an adsorbent's suitability for volatile species sequestration. Some of these parameters include (in no particular order) regenerability, chemical stability, mechanical stability, selectivity (i.e., rejection of non-target species), radiation stability, thermal stability, reactivity, density, and tolerance to process upset conditions. Capacity, density, mass transfer properties, and the needed decontamination factor are all important in determining the size of the bed required. The ease of converting used adsorber material to stable waste may also contribute to adsorbent selection.

Sequestration of volatiles and semivolatiles from reprocessing effluents by adsorption often must contend with several complicating factors. First, an adsorbent can also remove molecules other than the target from the gas stream, such as volatile components or water vapor. This often reduces the effective capacity of the adsorbent for the target volatile, and the phenomenon is sometimes termed *co-adsorption*. A prominent example of this is tritiated water vapor being co-adsorbed by silver mordenite, a well-studied sorbent considered for use in iodine capture [40]. Beyond effects on capacity, co-adsorption can alter mass transfer properties of the sorbent and decrease sorbent selectivity. Co-adsorption can also translate to effects on waste management, as co-adsorption of long-lived or radiotoxic species could increase the waste classification requirements of the adsorbate intended for short-lived, low toxicity species (e.g., iodine contamination of a tritium waste stream). Similarly, if 80% of the adsorption capacity is consumed by a non-target volatile, then the waste volume to be managed would increase correspondingly due to the increased amount of sorbent required to capture the same amount of target. For this reason, off-gas treatment systems should be designed to minimize co-adsorption of certain volatiles and off-gas treatment systems often contain many specialized capture processes with specialized adsorption materials.

A second complication in application of adsorption technology in treating effluents from nuclear fuel reprocessing is that the speciation and concentration of the volatiles can vary in different effluent streams. As an example, the target volatile species is likely to exist at a high concentration in a fuel pretreatment step like voloxidation but is likely to exist in other streams (like the COG stream) in very dilute concentrations (flow rates can also vary widely throughout the reprocessing facility). This can translate to different performance within different capture processes for the same adsorbent in the same facility. Selection of an adsorbent should take process conditions into consideration and ensure that any decrease in performance is addressed through either process design (where possible) or through using different adsorbents if needed.

Third, reprocessing techniques vary significantly. The most obvious is the difference between aqueous reprocessing (i.e., PUREX technology) and pyrochemical reprocessing (i.e., molten salt technology). However, even within these two approaches, there are substantial variances in potential implementation. For example, the addition of a fuel pretreatment step in either aqueous or pyrochemical reprocessing could result in new off-gas stream compositions and could necessitate different adsorbers. This complexity, especially when combined with issues such as co-adsorption and varying effluent composition, contributes to a complex treatment system in which many different operations will have to be designed and qualified for safe operations.

8.3. Sorption Applications

Adsorption is a desirable sequestration technology in the treatment of radiological gaseous effluents as it does not generate high-volume secondary liquid waste streams. It is therefore applied throughout the off-gas treatment systems where possible and is an area of active research and development. Adsorption using various adsorbents has been proposed for iodine, tritium (as both tritium and tritiated water), hydrochloric acid, noble gas management, and more. The most common applications for adsorption in reprocessing off-gas management are discussed here, but an exhaustive review is outside the scope of this document.

Iodine

Iodine may be present as elemental (I_2 gas), inorganic, or organic compounds within a reprocessing facility, and the specific forms will depend on the separations processes employed. Current practice in industrial reprocessing facilities (specifically, in the aqueous-based La Hague and Rokkasho facilities) is to use mineral-based adsorbents for iodine sequestration from the DOG stream. High-surface-area zeolite frameworks (most commonly, faujasite and mordenite-type zeolites) are impregnated with silver, which chemisorbs iodine as silver iodide (AgI) [41]. These materials have comparatively high iodine capacity, and the stable AgI compound is suitable as a secondary waste. The silver present in these minerals (i.e., AgX for faujasite, AgZ for mordenite) oxidizes in the DOG stream due to the high NO_x concentration, reducing its capacity by up to 80% as compared to ideal performance [42]. In a similar sorption mechanism, historical adsorbents were fabricated by mounting silver nitrate ($Ag\text{-}NO_3$) onto high-surface-area non-reactive alumina substrates. It has typically been assumed that the zeolite framework has improved acid resistance and limits the oxidation of the impregnated silver as compared to the simple $AgNO_3$-alumina concept.

A substantial amount of research and development has been performed on alternatives to silver-based sorbents for iodine capture. These efforts have included impregnated charcoals that are minerals with other active metals (Bi, Cu, Pb, etc.), MOFs, and silica materials [43]. In the United States, iodine release is expected to require very high levels of mitigation, and some scenarios suggest using solid adsorption in addition to a bulk iodine removal operation through wet scrubbing [44].

Iodine removal by sorbents is complicated by three factors. The first is that iodine may be present in multiple forms (i.e., elemental, organic, and inorganic compounds). Organic forms of iodine are more challenging to remove using traditional adsorption materials. The second is that in some reprocessing flowsheets, iodine concentration varies dramatically. Iodine in standard aqueous reprocessing flowsheet (no fuel pretreatment) partitions heavily to the DOG (at ppm concentrations) but also to the VOG and other streams (at ppb concentrations). Significant research has been devoted to obtaining sufficient data to design iodine adsorption operations for these highly dilute streams [45].

Tritiated Water

Depending on the specific reprocessing flowsheet that is implemented, tritium may partition to the off-gas as tritiated water. Molecular sieves, aluminosilicate minerals possessing pores of known size, are the most likely adsorbent material for use in tritiated water removal from reprocessing off-gas streams. Molecular sieves, often referred to as 3A-MS, 4A-MS, and 5A-MS for pore diameters of 3, 4, or 5 angstroms (Å), respectively, are robust and flexible, allowing their use in a variety of off-gas compositions. They are regenerable, with water captured at ambient temperatures and then desorbed at higher temperatures into smaller secondary waste streams for condensation and subsequent immobilization. The selectivity of molecular sieves is only based on the size of the sorbate. This limits their selectivity in that molecular sieves are likely to absorb other volatile species of similar size. This limitation is typically addressed by design choices. Molecular sieves have decreased capture efficiency in highly dilute streams, which has led in some cases to the deliberate addition of non-tritiated water to effect better overall separation factors for tritiated water [46].

Krypton/Xenon

As an alternative to cryogenic distillation, many different materials have been investigated for their use in adsorbing noble gases from off-gas streams. The most extensively researched materials are zeolites such as hydrogen mordenite (HZ) and silver-exchanged mordenite (AgZ), and MOFs. The ideal sorbent(s) would allow for separation of the two noble gases, concentration of the noble gases by adsorbing and then desorbing into a low-volume sweep gas, with all operational steps occurring at near atmospheric pressure and ambient temperature.

As an example, consider the conceptual implementation of HZ and AgZ to capture and separate Kr and Xe from reprocessing off-gas streams. A substantial amount of research and development has been performed in the United States to investigate this concept. A column of AgZ-PAN (PAN being a binder material for support) is contacted with the feed gas containing both Kr and Xe. The AgZ-PAN sorbent has an affinity for Xe at room temperature, and the effluent from the column will only contain Kr. This Kr-bearing stream is fed into an HZ-PAN column, which has an affinity for Kr at a temperature of 191 K, and the effluent from this column will not contain volatile radioactive species. The columns, once their capacity for each noble gas has been reached, will be rotated offline, and the noble gases will be desorbed by passing a sweep gas through the column at elevated temperatures, with the product being the two purified noble gases. Kr, which is radioactive, requires storage or disposal. In this scenario, chillers and heaters are critical to effective operation.

8.4. Wet Scrubbing Applications

Wet (or aqueous) scrubbing is an industrially common technology for removing pollutants, contaminants, or odor-causing compounds from process gas streams. Wet scrubbers can be purchased in fixed configurations from manufacturers, or they can be designed and fabricated for custom applications with straightforward reference criteria. Equipment is sized according to the velocity of the gas stream to be treated, the residence time needed for effective removal, and the size of any needed reservoir. An example of a wet scrubber system is shown in Figure 27.

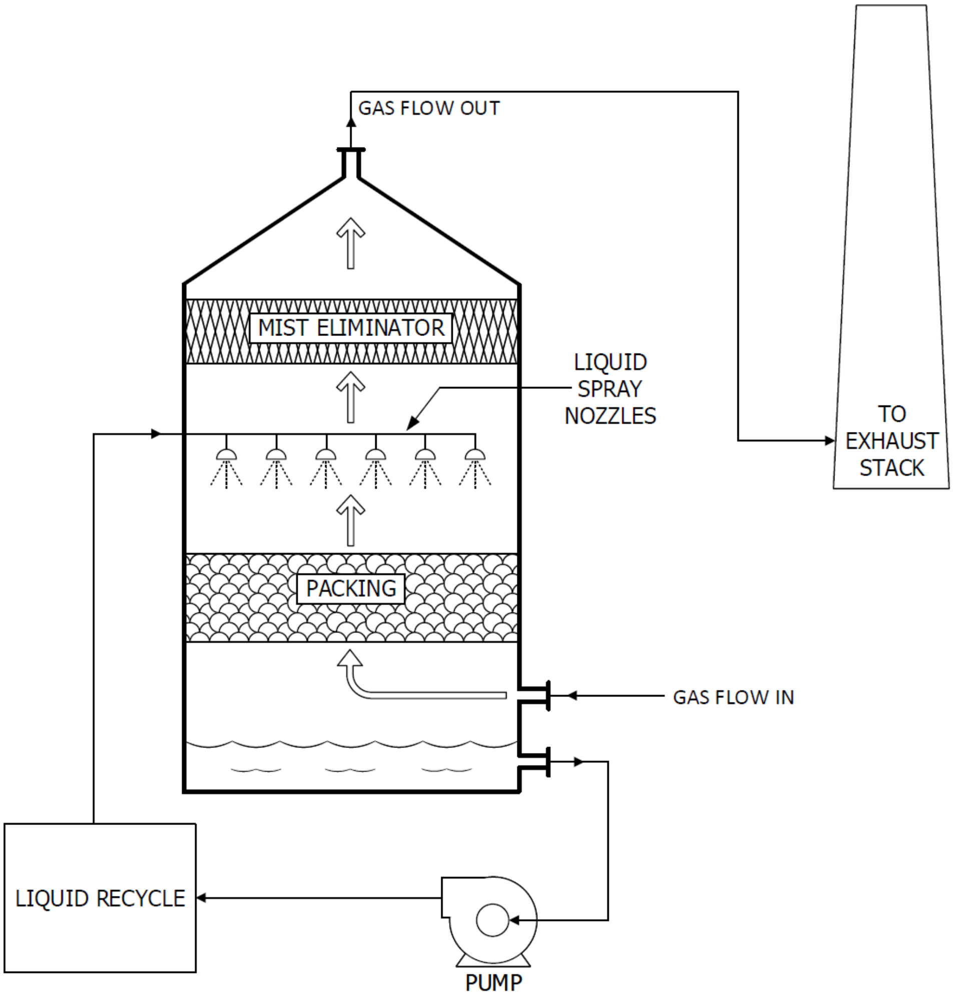

Figure 27. Schematic diagram of a wet scrubbing system for an off-gas stream.

Fundamentally, wet scrubbing works by contacting a flowing gas stream with a counter-current liquid. The liquid is selected for its affinity to remove the target contaminant from the gas stream. For instance, hydrochloric acid (HCl) gas can be removed by using a sodium hydroxide (NaOH) scrubber where the acid-base reaction between HCl and NaOH forms water and NaCl salt. The efficiency of a scrubber is highly dependent on the contact between the gas and liquid; maximizing the surface area of contact through generation of small gas bubbles or by using engineered packing materials is common. Residence time (the duration of contact between the gas and the scrubbing liquid) is also important to ensure effective transfer of the contaminant to the liquid phase.

Beyond the choice of scrubbing liquid, there are several key design choices that impact scrubber performance. First, increased scrubber tower height will increase residence time and increase separations efficiency. Second, scrubbers may use engineered packing materials to improve gas dispersion and increase surface area of contact between gas and liquid. These packing materials typically improve separations but are not indicated in cases where solid particulates or reaction byproducts could build up or plug the scrubber tower. In such cases, spray tower scrubbers (where liquid is sprayed onto the flowing gas stream at multiple points) are more desirable. Third, the velocity of the gas stream and the associated liquid:gas (L:G) flowrate ratio will affect separations. A lower gas velocity and a high L:G flowrate ratio will improve residence time and scrubber performance.

All scrubbers should include a mist eliminator on the gas stream prior to discharge. Mist elimination will ensure that the gas stream does not carry fine liquid droplets into the downstream ventilation system.

Wet scrubbers may be used in several different applications within a reprocessing off-gas treatment system. In nuclear applications, special considerations (such as the potential for accumulation of fissile material in a reservoir tank) must be taken to ensure the safety of the system. One benefit of wet scrubbers is that the reservoir tank often lends itself to continuous monitoring (e.g., by pH), which can enable very precise scrubber operation and reagent metering.

NO_x, iodine, and CO_2 are all candidates for removal by wet scrubbers. In the case of NO_x scrubbing, this is a more complex scrubbing technology with several potential design choices.

If wet scrubbing is implemented for iodine removal in a reprocessing facility, it may be used as the sole abatement technology or it may be used as a precursor to a solid adsorbent column. In the second scenario, the wet scrubber will remove the bulk of the iodine from the gas stream and the solid adsorbent can be used to further decrease the iodine concentration. Combining the two techniques may pose advantages, such as more complete removal of organic iodine species (these have limited retention by wet scrubbers), decreased solid sorbent handling and waste generation, and lower overall iodine concentrations in the effluent stream.

A caustic compound such as sodium hydroxide (NaOH) is the most likely solution to be used for wet scrubbing of iodine. Sodium hydroxide is also used elsewhere for iodine retention, such as in the wet seal of the DOG within the La Hague reprocessing facility. In the United States, the bench scale testing of iodine removal with a wet scrubber was recently demonstrated [44]. The iodine-bearing liquid waste from the scrubber will be an important secondary waste stream that will require immobilization and disposition.

CO_2 removal from off-gas streams may be pursued, although CO_2 is a minor contributor to the overall radiological dose arising from gaseous effluents of a reprocessing facility. CO_2 removal (sequestration) is an area of active research internationally, and the eventual design of a reprocessing facility may incorporate emerging technology for CO_2 capture. Historically, CO_2 removal has been proposed to proceed by wet scrubbing. A caustic solution (often NaOH) will react with CO_2 to form carbonate species such as sodium carbonate (Na_2CO_3). The carbonate species can be further reacted with alkaline metals (e.g., calcium hydroxide, $Ca(OH)_2$) to produce insoluble carbonates such as calcium carbonate ($CaCO_3$). This solid can then be filtered for immobilization and disposition.

9. AQUEOUS VS. PYROCHEMICAL OFF-GAS BEHAVIOR

The difference in technology readiness cannot be overstated. Aqueous reprocessing is a mature technology that has been used at industrial scales to support nuclear fuel cycles since the 1950s. By comparison, pyrochemical reprocessing has never been applied to support a nuclear fuel cycle, and off-gas capture technologies for pyrochemical reprocessing have never been applied for regulatory purposes.

Table 3 provides a side-by-side comparison of aqueous and pyrochemical reprocessing as a summary of information that was presented earlier in the report.

Table 3. Comparison between aqueous and pyrochemical off-gas behavior.

	Aqueous Reprocessing	Pyrochemical Reprocessing
1	Mature technology that has been demonstrated at industrial scales.	Early technology that has been demonstrated at research and engineering scales.
2	Relatively larger commercial scale plant.	Relatively smaller commercial scale plant.
3	Based on 40 CFR 190, emission may be controlled by more restrictive dose-based limits.	Based on 40 CFR 190, emission may be controlled by less restrictive fuel-cycle-based limits.
4	Off-gas capture technologies applied and demonstrated (in other countries) to meet emissions regulations.	Off-gas capture technologies not applied and demonstrated to meet emissions regulations.
5	Voloxidation may be used as a head-end process in both flowsheets. If applied, the majority of the volatile fission products will be released during decladding and voloxidation.	
6	Majority of volatile fission products are released during cladding breeching and fuel dissolution.	Majority of volatile fission products are released during cladding breeching, oxide reduction, and electrorefining.
7	Aqueous and organic solutions at temperatures from 25 to 70°C.	No aqueous or organic solutions. Except for the sodium wash station.
8	Calcination of actinide precipitates to form oxides at temperatures from 400 to 900°C	Molten chloride salts at temperatures from 500 to 650°C. Salt distillation and metal casting at temperatures from 800 to 1,500°C.
9	Off-gas includes vapors of water, acidic solutions, and organic solvents that condense upon cooling and require wet scrubbing.	Off-gas includes volatile metal oxides and metal chlorides that condense upon cooling and require filtration.
10	Iodine is released to the off-gas.	Iodine is captured in the chloride salts, but the extent of capture has not been validated as sufficient to meet emissions requirements.
11	Carbon is released to the off-gas.	Carbon is captured in the chloride salts, but the extent of capture has not been validated as sufficient to meet emissions requirements.

	Aqueous Reprocessing	Pyrochemical Reprocessing
12	Tritium follows the aqueous solutions and organic solvents and is released into the off-gas. Tritium spreads throughout the entire process as tritiated water.	Tritium is released to the off-gas with varied chemical speciation.
13	Noble gas fission products such as krypton-85 and xenon are released to the off-gas.	
14	The bulk of the off-gas may be air.	The bulk of the off-gas may be argon.

It may be the case that aqueous and pyrochemical reprocessing facilities will share similar capture technologies for iodine-129, krypton-85, carbon-14, and tritium. However, a significant difference is that an aqueous reprocessing facility will have a “wet” off-gas comprised mostly of air, while a pyrochemical reprocessing facility will have a “dry” off-gas comprised mostly of argon. The former may require wet scrubbing and filtration, while the latter may require only filtration.

Aqueous reprocessing includes high-temperature calcination operations to convert actinides precipitated from aqueous solutions into actinide oxides. However, volatile fission products have been removed prior to these calcination operations. This is not unlike some of the high-temperature salt distillation and metal casting operations used in pyrochemical reprocessing, where the volatile fission products are no longer present in the materials being processed.

10. APPLICABLE REGULATORY FRAMEWORK

This section provides a summary of the existing regulatory framework related to nuclear fuel reprocessing. 40 CFR 190 jointly considers the energy produced by the fuel cycle and the dose to surrounding populations. The derived regulatory discharge limits in 40 CFR 190 dictate the minimum capture efficiency requirements that the reprocessing facility must achieve for volatile radionuclides, thereby affecting the fundamental design characteristics of the off-gas ventilation systems. Given the importance of this regulation to the design requirements of a future reprocessing facility, 40 CFR 190 is reviewed here in depth. The regulatory guides provide an approved method to meet certain requirements in 10 CFR 50 and 10 CFR 20; therefore, these parts are also considered applicable to the design and operation of reprocessing facilities, and a brief discussion is provided on each. Reprocessing may also be licensed under 10 CFR Part 70; however, review of the applicable regulations was outside the scope of this report.

10.1. 40 CFR 190

10.1.1. 40 CFR 190: Overview

One of the most significant regulatory changes since RGs 3.20 and 3.32 were issued was the promulgation of the EPA regulation 40 CFR 190, “Environmental Radiation Protection Requirements for Normal Operations of Activities in the Uranium Fuel Cycle.” The 40 CFR 190 is a prescriptive regulation, with emissions from the entire fuel cycle restricted by both explicit quantities tied to electricity generation, e.g., mCi/GWy(e), and also whole body and thyroid doses that may be experienced by a member of the general public. It states:

> The annual dose equivalent does not exceed 25 millirems to the whole body, 75 millirems to the thyroid, and 25 millirems to any other organ of any member of the public as the result of exposures to planned discharges of radioactive materials, radon and its daughters excepted, to the general environment from uranium fuel cycle operations and to radiation from these operations…

and

> The total quantity of radioactive materials entering the general environment from the entire uranium fuel cycle, per gigawatt-y of electrical energy produced by the fuel cycle, contains less than 50,000 curies of krypton-85, 5 millicuries of iodine-129, and 0.5 millicuries combined of plutonium-239 and other alpha-emitting transuranic radionuclides with half-lives greater than one year.

Volatile emissions from an NRC-licensed reprocessing facility sited in the United States are expected to be regulated by both the NRC and EPA through 10 CFR 20 [47] and 40 CFR 190 [48]. Jubin et al. [49] provided a comprehensive analysis of how these regulations might apply to an aqueous separations-based reprocessing facility, and that analysis was extended to a pyrochemical-based reprocessing facility by Bruffey et al. [50]. Key observations from these reports relevant to the current discussion are presented here, along with additional observation and analysis.

10.1.2. 40 CFR 190: Fuel Cycle Limits on Volatile Release

The most explicit language in 40 CFR 190 is that of the specific curie (Ci) limits listed for Kr-85, I-129, and Pu-239 (and other alpha-emitting transuranics). The release limits prescribed in 40 CFR 190 are summarized in Table 4 and converted to their mass equivalents (based on the specific activity of each isotope [51]) for ease of reference. **These Ci release limits are tied to the energy produced by the entire fuel cycle, which thus requires that off-gas emissions must be evaluated in the context of the entire fuel cycle, and not just in the context of a single reprocessing facility.**

Table 4. Environmental release limits of select isotopes per GWe-y of electricity production.

Isotope	Limit (MBq)	Limit (Ci)	Specific Activity (Ci/g)	Mass Equivalent (g)
Kr-85	1,850,000,000	50,000	390	128.2
I-129	185	0.005	0.00018	27.8
Pu-239	18.5	0.0005	0.062	0.00806
Note: Data for Pu-239 is provided as an example. Actual limit includes all alpha-emitting transuranic radionuclides with half-lives greater than 1 year.				

In 2022, the United States produced 804 TWe-hr of electrical energy from nuclear reactors [52], which is equivalent to 91.8 GWe-y as shown below.

$$804\,TWe\text{-}hr \cdot \frac{d}{24\,hr} \cdot \frac{y}{365\,d} \cdot \frac{1000\,GWe}{TWe} = 91.8\,GWe\text{-}y$$

Based on this conversion from 1 GWe-y (per the EPA regulation) to 91.8 GWe-y (per the annual nuclear electrical energy production in the United States), the environmental release limits of Kr-85, I-129, and all alpha-emitting transuranic radionuclides with half-lives greater than 1 year are summarized in Table 5. The assumption is that the EPA regulation is meant to scale with U.S. domestic energy production in this way.

Table 5. Environmental release limits of select isotopes per 91.8 GWe-y of electricity production.

Isotope	Limit (MBq)	Limit (Ci)	Specific Activity (Ci/g)	Mass Equivalent (g)
Kr-85	169,830,000,000	4,590,000	390	11,769
I-129	16,983	0.459	0.00018	2,552
Pu-239	1,698	0.0459	0.062	0.740
Note: Data for Pu-239 is provided as an example. Actual limit includes all alpha-emitting transuranic radionuclides with half-lives greater than 1 year.				

At standard temperature and pressure conditions of 0°C and 1 bar, respectively, 11.769 kg of Kr-85 gas would occupy a volume of 3.14 m^3, and 2.552 kg of I-129 gas would occupy a volume of 0.23 m^3.

The examples that follow are not demonstrations of rigorous calculations. They are meant to illustrate how the environmental release limits and capture efficiencies of a reprocessing facility are impacted by factors such as fuel burnup and reactor thermal-to-electrical conversion efficiency. It is understood that the regulations outlined in EPA 40 CFR 190 are tied to the energy produced by the entire fuel cycle.

10.1.3. 40 CFR 190: Fuel Cycle Limits to Reprocessing Light-Water Reactor Fuel

Typical LWR fuel is uranium oxide (UO_2) clad in a zirconium alloy. The burnup of the spent LWR fuel is commonly measured in GWt-d per MT of uranium. The thermal energy to electrical energy conversion efficiency of an LWR is on the order of 33%. For example, an LWR that produces 3,000 MWt of thermal energy will generate 1,000 MWe of electrical energy. The regulatory statement is based on "gigawatt-year of electricity produced." Concentrations of Kr-85 and I-129 in LWR uranium oxide fuel as a function of burnup in units of GWt-d per MT of uranium are shown in Figure 28. The two sets of data were fit to linear regression equations, which are shown in the figure. The data in Figure 28 are previously unpublished and were derived from the modeled analysis of actual spent fuel from a thermal neutron spectrum LWR. Modeling was performed using Oak Ridge Isotope Generation (ORIGEN)-based burnup code [53].

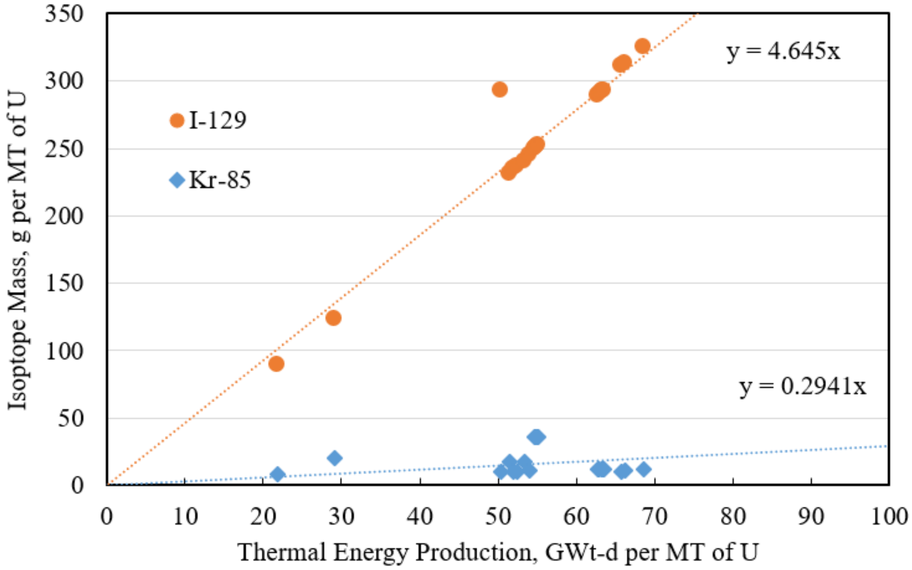

Figure 28. Modeled concentrations of select isotopes in LWR uranium oxide fuel as a function of thermal energy production (GWt-d per MT of U).

Based on a thermal-to-electrical energy conversion efficiency of 33%, the thermal energy production data in units of GWt-d per MT of uranium shown in Figure 28 are converted to electrical energy production data in units of GWe-d per MT of uranium in Figure 29.

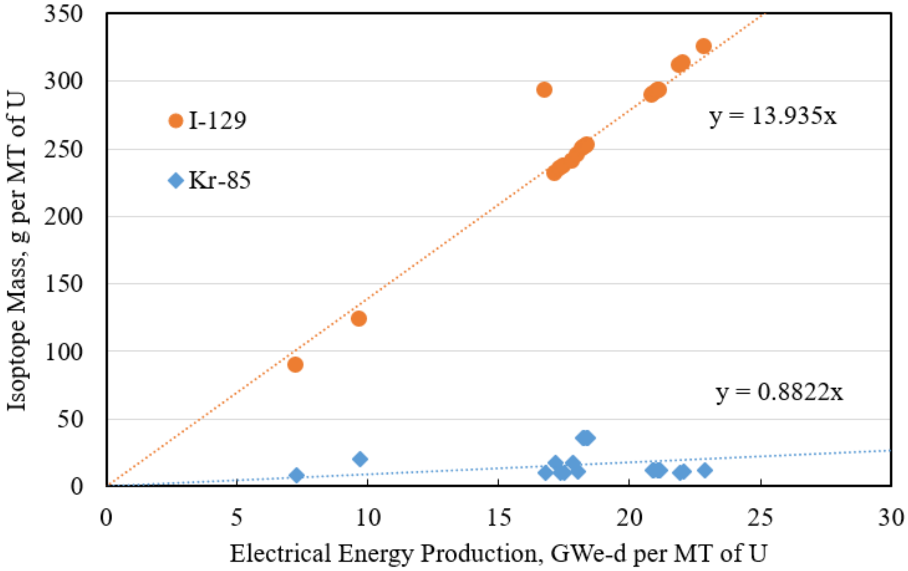

Figure 29. Modeled concentrations of select isotopes in LWR uranium oxide fuel as a function of electrical energy production (GWe-d per MT of U).

The electrical energy production data in units of GWe-d per MT of uranium in Figure 29 are converted to units of GWe-y per MT of uranium in Figure 30.

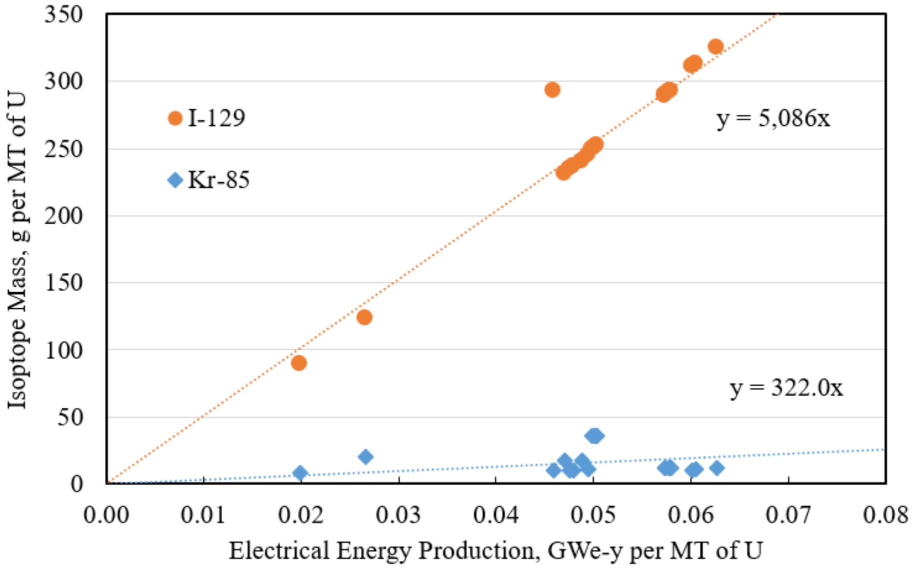

Figure 30. Modeled concentrations of select isotopes in LWR uranium oxide fuel as a function of electrical energy production (GWe-y per MT of U).

A metric ton of uranium (U) is contained in 1.134 MT of uranium oxide (UO_2). The electrical energy production data in units of GWe-y per MT of uranium in Figure 30 are converted to units of GWe-y per MT of uranium oxide in Figure 31.

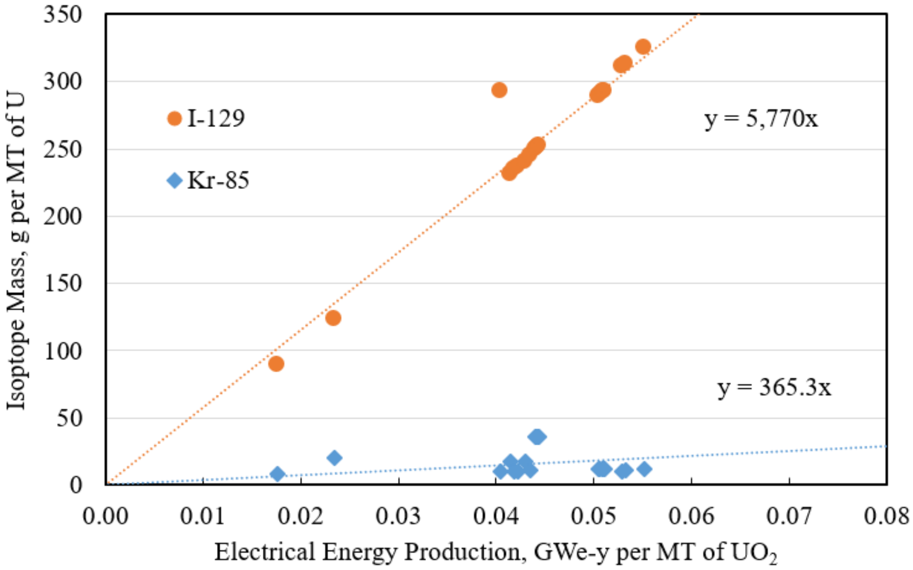

Figure 31. Modeled concentrations of select isotopes in LWR uranium oxide fuel as a function of electrical energy production (GWe-y per MT of UO_2).

Based on this modeled data for LWR uranium oxide fuel, a burnup of 60 GWt-d per MT of uranium is equivalent to 20 GWe-d per MT of uranium, which is equivalent to 0.0548 GWe-y per MT of uranium, in turn, to 0.0483 GWe-y per MT of uranium oxide. These conversions are summarized in Table 6.

Table 6. Summary of energy unit conversions.

Units	Method of Conversion
60 GWt-d per MT of uranium	Original Modeled Data
20 GWe-d per MT of uranium	Convert Thermal to Electrical
0.05479 GWe-y per MT of uranium	Convert Day to Year
0.04832 GWe-y per MT of uranium oxide	Convert Uranium to Uranium Oxide (UO_2)

A reprocessing facility operating at a capacity of 100 MT per year of LWR uranium oxide fuel would be equivalent to 4.832 GWe-y of "electricity production."

Based on the conversion from 1 GWe-y (per the EPA regulation) to 4.832 GWe-y (per the reprocessing capacity of 100 MT per year), the environmental release limits of Kr-85, I-129 and all alpha-emitting transuranic radionuclides with half-lives greater than 1 year are summarized in Table 7. These values are applicable to the entire fuel cycle—if emissions occur in other fuel cycle operations, the sum of all emissions across the fuel cycle must be within these limits for the reference scenario.

Table 7. Environmental release limits of select isotopes per 100 MT of LWR uranium oxide fuel.

Isotope	Limit (MBq)	Limit (Ci)	Specific Activity (Ci/g)	Mass Equivalent (g)
Kr-85	8,939,200,000	241,600	390	619.5
I-129	893.92	0.02416	0.00018	134.2
Pu-239	89.392	0.002416	0.062	0.03897
Note: Data for Pu-239 is provided as an example. Actual limit includes all alpha-emitting transuranic radionuclides with half-lives greater than 1 year.				

Based on a fuel burnup of 0.04832 GWe-y per MT of uranium oxide and the data in Figure 31, 100 MT of spent LWR fuel contains 27,870 g of I-129 and 1,764 g of Kr-85.

The necessary capture efficiency of Kr-85 is 64.9% as shown below.

$$\left(1-\frac{619.5\ g\ Kr\text{-}85\ Released}{1{,}764\ g\ Kr\text{-}85\ Present}\right)\cdot 100 = 64.9\%$$

The necessary capture efficiency of I-129 is 99.5% as shown below.

$$\left(1-\frac{134.2\ g\ I\text{-}129\ Released}{27{,}780\ g\ I\text{-}129\ Present}\right)\cdot 100 = 99.5\%$$

10.1.4. 40 CFR 190: Fuel Cycle Limits to Reprocessing Sodium-Cooled Fast Reactor Fuel

Typical SFR fuel is a metallic alloy of uranium and zirconium clad in stainless steel. Modeled concentrations of Kr-85 and I-129 in SFR metallic fuel as a function of burnup in units of GWt-d per MT of uranium are shown in Figure 32. The two sets of data were fit to linear regression equations, which are shown in the figure. The data in Figure 32 are previously unpublished and were derived from the modeled analysis of actual spent fuel from a fast neutron spectrum SFR. Modeling was performed using ORIGEN-based burnup code. The units of atomic percent were converted to units of GWt-d per MT of uranium based on 6.5 atomic percent being equivalent to 60 GWt-d per MT of uranium. Notice that the data in Figure 32 for SFR fuel is very similar to the data in Figure 28 for LWR fuel.

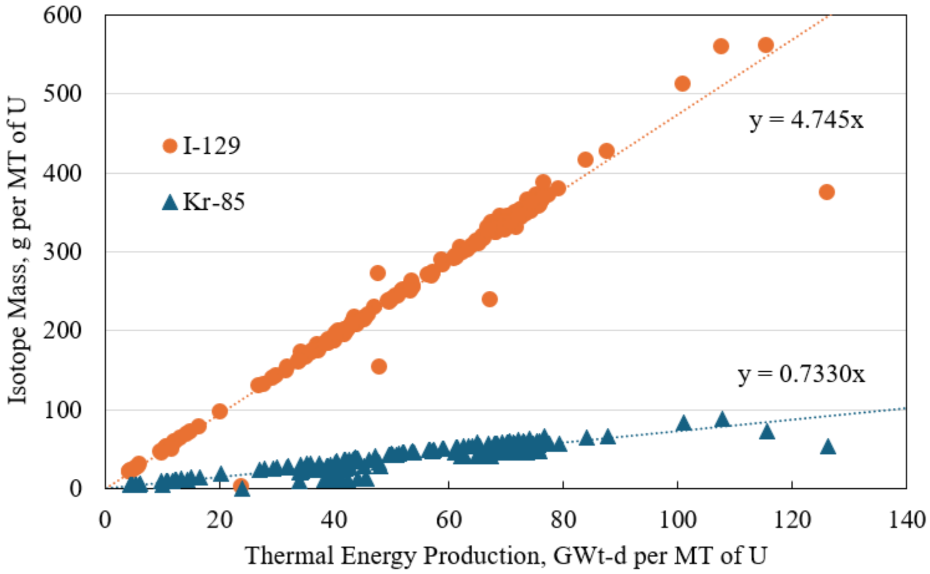

Figure 32. Modeled concentrations of select isotopes in SFR metallic fuel as a function of thermal energy production (GWt-d per MT of U).

The thermal energy production data in units of GWt-d per MT of uranium shown in Figure 32 are converted to electrical energy production data in units of GWe-y per MT of uranium in Figure 33. The data in Figure 33 for the SFR example reflect a thermal energy to electrical energy conversion efficiency of 33%, which was also used for the LWR example.

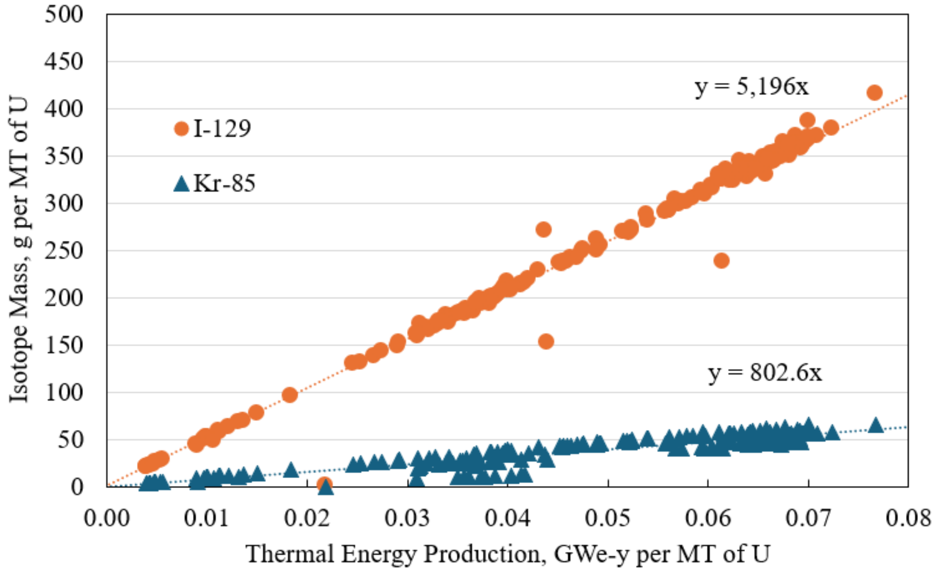

Figure 33. Modeled concentrations of select isotopes in SFR metallic fuel as a function of thermal energy production (GWe-y per MT of U).

A reprocessing facility operating at a capacity of 100 MT per year of SFR metallic uranium fuel would be equivalent to 5.479 GWe-y of "electricity production."

Based on the conversion from 1 GWe-y (per the EPA regulation) to 5.479 GWe-y (per the reprocessing capacity of 100 MT per year), the environmental release limits of Kr-85, I-129 and all alpha-emitting transuranic radionuclides with half-lives greater than 1 year are summarized in Table 8.

Table 8. Environmental release limits of select isotopes per 100 MT of SFR metallic fuel.

Isotope	Limit (MBq)	Limit (Ci)	Specific Activity (Ci/g)	Mass Equivalent (g)
Kr-85	10,136,150,000	273,950	390	702.4
I-129	1,013.62	0.02740	0.00018	152.2
Pu-239	101.362	0.002740	0.062	0.04419
Note: Data for Pu-239 is provided as an example. Actual limit includes all alpha-emitting transuranic radionuclides with half-lives greater than one year.				

Based on a fuel burnup of 0.05479 GWe-y per MT of uranium and the data in Figure 33, 100 MT of spent SFR fuel contains 28,469 g of I-129 and 4,397 g of Kr-85.

The necessary capture efficiency of Kr-85 is 64.9% as shown below.

$$\left(1-\frac{702.4\ g\ Kr\text{-}85\ Released}{4{,}397\ g\ Kr\text{-}85\ Present}\right)\cdot 100 = 84.0\%$$

The necessary capture efficiency of I-129 is 99.5% as shown below.

$$\left(1-\frac{152.2\ g\ I\text{-}129\ Released}{28{,}469\ g\ I\text{-}129\ Present}\right)\cdot 100 = 99.5\%$$

10.1.5. 40 CFR 190: Comments on Fission Yield

Fission yield curves of nuclear fuels exhibit characteristic bimodal distributions. Two maxima peaks occur in the vicinity of atomic masses of 90–100 and 130–140. Yield drops off rapidly below 90 and above 140. Between 100 and 130, there is a minima trough, which is influenced by the neutron energy in the core. Thermal spectrum neutrons lower the trough (i.e., lower yield in this region); fast spectrum neutrons raise the trough (i.e., higher yield in this region). Fission yield is influenced by factors such as the composition of the fuel, neutron spectrum, burnup, and age. However, from the perspective of reprocessing, the variations in fission yield from one reactor to another may be of little consequence to the emissions performance of an aqueous or pyrochemical reprocessing facility. The overriding concern is which reprocessing technology, aqueous or pyrochemical, is best suited for the given fuel cycle application.

The two examples given above were for LWR oxide fuel and SFR metallic fuel at similar burnups. In many ways, these are two very different reactors. Nevertheless, in many ways, the compositions of the spent fuels were similar as demonstrated by comparing the data in Figure 28 and Figure 32. The relative abundances of I-129 and K-85 are similar between the two fuel types.

10.1.6. 40 CFR 190: Summary of Fuel Cycle Limits

The evaluation presented here is consistent with the limits on volatile emissions arising from the fuel cycle limits prescribed in 40 CFR 190 and with the analyses performed by Jubin et al. [49] and Bruffey et al. [54]. For a facility with up to 200 MT per year processing throughput (a likely maximum for the throughput of a pyrochemical reprocessing facility), Bruffey et al. [54] showed that volatile emissions are likely to be governed by these fuel cycle limits. However, aqueous reprocessing facilities have historically been designed for much higher throughput, and when throughput is 1000 MT per year (or higher, as in the case of the La Hague facility in France), the total release of radioactive material may begin to significantly contribute to the public dose of a facility even if the fuel cycle limits are met. In these cases, the dose-based limits from the second part of 40 CFR 190 must be considered in order to achieve regulatory compliance. These limits are discussed in the next section.

10.1.7. 40 CFR 190: Dose-Based Limits on Volatile Releases

10 CFR 20 is applicable to NRC-licensed facilities and limits the dose to the public to less than 100 mrem per year. 10 CFR 20 allows the licensee to demonstrate compliance with this limit by either: (1) demonstrating through measurement or calculation that the dose to the public is below annual limits or (2) demonstrating that annual average concentrations for gaseous and liquid effluents are below certain values (provided in 10 CFR 20, Appendix B) at the site boundary.

Both 10 CFR 20 and 40 CFR 190 include dose-based limits that will have to take into account the cumulative dose to the public from multiple radionuclides arising from both liquid discharges and gaseous emissions. The dose limits prescribed by 40 CFR 190 are shown in Table 9 and can be compared to the 620 mrem annual average dose per person from all natural and manmade sources [55]. However, an operating facility will not measure this dose; it will instead quantitatively measure components of specific effluent streams prior to discharge, through either chemical or radiological measurements (e.g., gamma monitoring of a facility stack).

Table 9. Annual dose equivalent limits to population.

Human Exposure Location	Annual Dose Equivalent	
	(mrem)	(mSv)
Whole Body	25	0.25
Thyroid	75	0.75
Any Other Organ	25	0.25

The dose-based language used in the regulations gives rise to a condition in which the specific measured values assigned as administrative limits to a reprocessing facility's gaseous emissions arise from a balanced trade-off between numerous variables. For example, if the total dose arising from liquid discharges is extremely low, then additional flexibility can be gained in the release of volatile species. Similarly, if long-cooled fuel is processed, then almost no controls are required for Kr-85 emissions, and additional margin is available for dose contributions from iodine. In all cases, the facility will need to keep emissions at or below the values required for regulatory compliance. NRC issued guidance on designing an acceptable program for establishing and maintaining as low as reasonably achievable (ALARA) levels for gaseous and liquid effluents at materials facilities. Though not directly applicable to reprocessing facilities, an applicant may want to consider this guidance when designing their off-gas ventilations systems [56].

Given the complexity of balancing these variables, identifying specific release values or needed in facility decontamination factors for volatile species is challenging and prevents the more straightforward analysis presented for the 40 CFR 190 fuel cycle limits in Section 10.1.3. It requires making numerous assumptions about plant design and operation. It also requires performing assessments of environmental dispersion to identify how gaseous emissions would distribute into the atmosphere, thereby eventually providing dose to members of the public outside of the site boundary. Thus, the efforts described in Jubin et al. [49] and Bruffey et al. [54] were meant to provide an assessment of a generalized reprocessing facility such that a broad envelope for needed decontamination factors could be established. Analysis of an actual facility would allow the assumptions on siting, design, and operation to be replaced with true values that facilitate a more detailed understanding of specific plant requirements. In general, both aqueous and pyrochemical assessments showed that I-129 would be subject to the strictest controls of the four primary volatile species. The envelope for potential I-129 decontamination factors was generally observed to be in the thousands (with substantial variance across the scenarios studied). Tritium was found to require maximum decontamination factors in the hundreds with some cases requiring no abatement (with the lowest values observed for long-cooled fuels and low facility throughputs). Decontamination factors calculated for Kr-85 were below 100, again requiring no mitigation in some scenarios. Carbon-14 only required mitigation in the most restrictive scenarios.

A non-exhaustive list of variables that affect required decontamination factors includes the following:

- Geographical location and associated meteorological conditions of the facility
- Physical footprint of the facility, including the distance from the discharge stack to the public site boundary
- Design basis of facility, including the rated throughput, selected chemical processing method, and the incorporation of any recycle loops for aqueous or gaseous process streams
- Characteristics of the processed fuel, including burnup fraction and cooling time.

Atmospheric discharge limits of radionuclides from a reprocessing facility are regulated by 40 CFR 190 by joint considerations to the energy produced by the fuel cycle and the dose to surrounding populations. In turn, these discharge limits dictate the minimum capture efficiency requirements that the reprocessing facility must achieve, thereby affecting the fundamental design characteristics of the off-gas ventilation systems.

10.2. 10 CFR 50

10.2.1. 10 CFR 50: Domestic Licensing of Production and Utilization Facilities

At the time the RGs were published, § 50.34, "Contents of Applications" discussed the need for an applicant as follows:

> (3) the preliminary design [criteria] of the facility including: (i) The principal design criteria for the facility…(ii) The design bases and the relation of the design bases to the principal design criteria.

These requirements in § 50.34 that pertain to fuel reprocessing facilities have remained the same since 1974.

Around the same time as the RGs were issued, the Atomic Energy Commission issued a Federal Register notice in 1974 (39 FR 26293) that detailed the consideration of amendments to add an Appendix P, "General Design Criteria for Fuel Reprocessing Plants" to 10 CFR 50, "Licensing of Production and Utilization Facilities" (as it was then titled). Though Appendix P was not promulgated, the draft General Design Criteria contained criterion specific to off-gas and ventilation systems as follows:

> The ventilation and off-gas systems shall be designed and tested to assure the confinement of radioactive materials during normal or abnormal conditions. To accomplish this objective, these systems shall be designed to meet the following requirements:
>
> a) The proper ventilating air flow direction shall be maintained across the confinement barrier, that is, between areas inside the barrier and areas outside the barrier, under operating and accident conditions.
>
> b) The ventilation system shall accommodate changes in operating conditions such as variations in temperature or pressure and shall be capable of safely

controlling all radioactive off-gases that could be associated with normal or accident conditions.

c) The continuity of necessary ventilation shall be ensured by means of redundant equipment, fail-safe control systems, or other provisions.

d) Provisions shall be made for testing all safety-related components during normal operation of the systems to demonstrate their ability to perform at design efficiency and to function during emergency conditions and during transitions between normal end emergency conditions.

e) Ventilation systems shall be designed to permit the continued occupancy of any and all areas where such occupancy is required for normal plant operation for safe shutdown or maintaining the plant in a safe shutdown condition. The design shall include protection against the intake or accumulation of radioactive materials. The design shall also permit the timely and safe execution of personnel from all areas.

f) Vessel and dissolver off-gas systems shall be designed to confine the radioactive materials during normal operation and to assure that the concentration of radioactive materials in the effluent gases is as low as practicable. Such systems shall also be designed to retain their confinement and separation capability to reduce releases resulting from an accident condition to levels consistent with the regulations contained in this chapter.

It should be noted that Appendix P to Part 50 has been reserved for future use in the NRC's regulatory framework.

10.2.2. 10 CFR 50, Appendix B: Quality Assurance Criteria for Nuclear Power Plants and Fuel Reprocessing Plants

RG 3.20 refers to the pertinent quality assurance requirements of Appendix B to Part 50 and how the regulation should be applied to all activities affecting the safety-related functions of these structures, systems, and components. It is noted that the text of this appendix has not changed significantly since 1974.

It can be concluded that the regulations, which both the off-gas and ventilation system RGs provide a method of how to meet, have not changed significantly since 1975.

10.3. 10 CFR 20

10.3.1. 10 CFR 20: Standards for Protection Against Radiation

Gaseous effluents released by a reprocessing facility would be regulated under 10 CFR 20, "Standards for Protection against Radiation," using standards similar to the U.S. EPA's National Emissions Standards for Hazardous Air Pollutants (NESHAP) regulations. Though the regulation is mentioned in RG 3.20, no specific details were provided in the RG on how to meet the requirements therein.

ALARA has been a central tenet of radiation protection for many years and is a requirement in many NRC regulations. However, when the RGs were issued, ALARA was not explicitly included in 10 CFR 20. However, the intent of ALARA was promulgated in § 20.1(c) "…in addition to complying with the requirements set forth in this part, make every reasonable effort to maintain radiation exposures, and releases of radioactive materials in effluents to unrestricted areas, as far below the limits specified in this part as practicable." The phrase "as far below the limits specified in this part as practicable" means as low as is practicably achievable taking into account the state of technology and the economics of improvements in relation to benefits to the public health and safety and in relation to the utilization of atomic energy in the public interest.

A major revision to 10 CFR 20 was promulgated in 1991 (56 FR 23360), which incorporated updated scientific information to reflect changes in the basic philosophy of radiation protection. However, since the statement pertaining to 10 CFR 20 in the RG is performance-based, changes to 10 CFR 20 since 1974 do not have an impact on the RG 3.20's applicability to 10 CFR 20. The RG states that "[a] parts of the process off-gas systems should be designed to…limit the release of radioactive materials during normal operation to the levels stated in 10 CFR Part 20." Similarly in draft RG 3.32, text pertaining to 10 CFR 20 is similarly written, "The systems must continue to perform their safety functions effectively under all conditions by confining radioactive or other potentially dangerous materials. They should be designed to ensure that the concentration of radioactive materials in the effluent gases is as far below the limits specified in 10 CFR 20 as practicable."

A reprocessing facility will have to meet the effluent limits established by EPA in 40 CFR 190. As per § 20.2203, "Reports of exposures, radiation levels, and concentrations of radioactive material exceeding the constraints or limits," subpart (4) states "[f]or licensees subject to the provisions of EPA's generally applicable environmental radiation standards in 40 CFR [P]art 190, levels of radiation or releases of radioactive material in excess of those standards, or of license conditions related to those standards." These limits apply to "uranium fuel cycle" operations, which include the "reprocessing of spent uranium fuel" (see 40 CFR 190.02). The requirements consist of two parts: a dose limit and a release limit. The NRC implements these standards in 10 CFR 20. The NRC, in collaboration with EPA, may want to consider developing an RG that would explain to licensees how to interpret the quantitative limits in 40 CFR 190 for reprocessing facilities.

11. SUMMARIES OF NRC REGULATORY GUIDES 3.18, 3.20, 3.32, AND 3.12

NRC RGs 3.18 (February 1974), 3.20 (February 1974), and 3.32 (September 1975), which address "Fuel Reprocessing Plants," were written for PUREX-based aqueous reprocessing of LWR uranium oxide fuels. RG 3.12 (December 2010), which addresses "Plutonium Processing and Fuel Fabrication Plants," was written for MOX fuel fabrication for LWRs. This section provides a summary of the salient design recommendations for ventilation systems made in these four RGs.

Both RGs 3.20 and 3.32 refer to § 50.34, "Contents of applications; technical information," specifically for providing guidance on how a reprocessing facility license can meet the requirements in Subsection (3) regarding the design of the facility. In the case of RG 3.20, the purpose of the guide is to provide information related to establishing principal design criteria for equipment associated with process off-gas treatment systems for fuel reprocessing plants. RG 3.32 describes a bases acceptable to the NRC staff for the design of ventilation systems for fuel reprocessing plants.

11.1. Summary of NRC RG 3.18

Confinement Barriers and Systems for Fuel Reprocessing Plants

- Confinement barriers and confinement systems:
    - "*Total* barriers" are those fabricated of impermeable materials expected to prevent penetration of all confined material without regard to that material's physical or chemical nature.
    - "*Selective barriers*" are mass transfer devices or filters employed to remove selected chemicals or particulate matter from a ventilation stream while allowing the bulk of the stream to pass through.
    - Confinement systems and barriers should limit the release of radioactive material to restricted areas so that exposures to individuals are as low as practicable and that the concentrations are as low as practicable.
    - Confinement systems should withstand the effects of design basis accidents so that releases of radioactive materials are as low as practicable.
    - Barriers should withstand loading due to pressure differentials imposed by process off-gas and building ventilation systems.
    - Openings through barriers should preclude disruption of confinement or disruption of air flow patterns due to loss of pressure differentials.
- Zones to prevent the spread of contamination:
    - Zones are separated by confinement barriers into areas of various levels of contamination.
    - Zones should be arranged in order of contamination potential. Zones with the highest contamination potential are adjacent to contaminated process areas. Zones with the lowest contamination potential are adjacent to the outer areas of the process building.
    - Air flows successively from zones of lower contamination potential to zones of higher contamination potential.
    - Air flows successively from the ambient surroundings, to building areas occupied by personnel, to potentially contaminated process areas, to the ventilation cleanup systems, and to blowers for discharge to the stack.
- The capacity of ventilation systems must:
    - The velocity of gas flowing through any barrier opening must be sufficient to prevent the backflow of airborne contaminants through such openings.
    - Air flow patterns must not be disrupted by winds, movement of equipment or personnel, or temporary opening of passageways through confinement barriers.

- Penetrations through confinement barriers:
  - Routing of fluids such as water, steam, and air provide pathways for the potential release of radioactive materials, especially when circulating between radioactive and nonradioactive areas.
  - Penetrations serve as conduits between zones formed by the barriers and provide potential pathways for the release of radioactive materials.
  - Penetrations should be fitted with means of isolating selected equipment to assure confinement during maintenance activities.
  - Pipes penetrating confinement barriers should be equipped with isolation valves located as close to the confinement barrier as is practicable.
- Monitoring devices:
  - Where a total barrier is used for gas confinement, integrity may be inferred by monitoring the difference in pressure across the barrier and noting any deviation from normal.
  - Where a total barrier is used for liquid confinement, leakage may be indicated by a liquid high-level alarm in a specially designated leakage collection sump, or by analytical devices which measure and alarm when contaminant intrudes into normally uncontaminated areas.
  - Monitors are used to indicate any breach in integrity of a total barrier or any loss of function of a selective barrier which would release radioactivity from a designated confinement area.

11.2. Summary of NRC RG 3.20

Process Off-gas Systems for Fuel Reprocessing Plants

- Usual constituents of process off-gas systems are:
  - Off-gas collection systems.
  - Condensate removal systems.
  - Sampling and chemical monitoring systems.
  - Control and instrumentation systems.
- Prevent uncontrolled releases of radioactive material under:
  - Normal operating loads.
  - Loads attributable to natural phenomena.
  - Utility failures.
  - Failures due to engineering or operating errors.
- Off-gas systems are generally equipped with:
  - Duplicate equipment items designed to maintain critical functions.
  - Spare exhaust blowers.
  - Equipment that will automatically switch to emergency utility systems.
- To assure system reliability the materials used in lines and equipment must be:
  - Nonflammable.
  - Resistant to heat.
  - Resistant to corrosive effects of the collected gases.
  - Resistant to strong chemicals used for equipment decontamination.
- Off-gas systems function to handle:
  - Condensable gases, primarily steam and water vapor.

  - Non-condensable gases.
  - Condensables removed by condensers and stack drains.
  - Sampling and monitoring of all effluents.
- Off-gas systems are treated by mass transfer devices:
  - Caustic scrubbers.
  - Acid scrubbers.
  - Activated carbon adsorbers.
  - Zeolite adsorbers.
  - Water scrubbers to remove oxides of nitrogen.
- Off-gas system filters should be:
  - Resistant to fire.
  - Preceded by fire-suppressing equipment.
  - Filters and adsorbers may be preceded by heaters.
  - Filters and absorbers may be preceded by electrical or steam traced lines.
- Ventilation and off-gas systems work together to:
  - Keep process vessel pressures lower than pressures in surrounding process.
  - Limit leakage of contaminated gases into process cells.
  - Limit leakage into potentially inhabited plant areas.

11.3. Summary of NRC RG 3.32

General Design Guide for Ventilation Systems for Fuel Reprocessing Plants

- General safety of ventilation systems:
  - Ventilation systems should be designed to prevent uncontrolled release and confine radioactive materials as close to the point of origin as practicable.
  - Confinement of radioactive materials should be provided by multiple-zone confinement barriers and systems.
  - Negative pressure differentials should be maintained to ensure that air flow is from zones of lower contamination potential to zones of higher contamination potential.
  - Ventilation systems should be designed so that the failure of any one component will not affect the continuous operation of the ventilation systems.
  - Ventilation systems should be capable of operating during normal power outage at capacities required to maintain confinement of contaminants.
  - Ventilation systems should be designed to withstand any credible fire and explosion and continue to act as confinement barriers.
  - Ventilation systems should be constructed of fire-resistant materials and designed to restrict the spread of fires.
  - Ventilation systems should be designed to withstand credible natural disaster conditions without loss of confinement capability.
- Occupied area ventilation systems:
  - Where air from occupied areas is directed to contaminated or potentially contaminated areas, consideration should be given to passing this air through suitable filters to prevent backflow of particulate contaminants.

  - Air locks should be provided where frequent entry between personnel occupancy areas and limited access areas is necessary and where air flow must be maintained in one direction.
- Process area ventilation systems:
  - The ventilation systems should maintain minimum air velocities of 120 linear feet per minute (design velocity should be ~150 linear feet per minute) through all process area openings to prevent significant reverse flow of contaminated air.
- Exhaust ventilation and filtration systems:
  - The filtered air should be discharged to the environment through a stack of sufficient height to reduce close-in ground-level concentrations of radioactive or other potentially dangerous contaminants.
  - Provisions should be made for an alternative release point in the event that off-gas flow through the stack is blocked because of structural failure.
  - The stack should be located sufficiently distant from other facilities such that structural failure would not result in damage to any process systems or structures important to safety.
- Ventilation system testing and monitoring:
  - Exhaust ducts and stacks that may contain radioactive contaminants should be provided with fixed samplers and continuous air monitors (CAMs)that provide a record of plant effluents.
  - Air monitoring and warning systems should be installed in areas where radioactive material is handled.

11.4. Summary of NRC RG 3.12

General Design Guide for Ventilation Systems of Plutonium Processing and Fuel Fabrication Plants

This RG refers to a facility that would be used to process weapons grade plutonium (WGPu) into mixed oxide (MOX) fuel for use in LWRs. This was the purpose of the MOX facility that was being constructed at the Savannah River Site but was ultimately abandoned. Historically, WGPu was produced in dedicated plutonium production reactors. The spent fuel from these reactors was processed to recover purified plutonium as PuF_4 (in the early years) and PuO_2 (in later years). In other words, the plutonium of concern to this RG is very clean material that is free of the fission products usually associated with spent fuels. It is plutonium that can be processed in unshielded gloveboxes. It is likely to be predominantly Pu-239, Pu-240, Pu-241, and Am-241. This limitation implies that no off-gas capture technologies are needed, other than filtration.

This RG uses the terms integrated safety analysis (ISA) and an item relied on for safety (IROFS). The findings of the ISA determine whether a mechanical system is, or is not, an IROFS. In broad terms, systems that are IROFS are those that prevent, or minimize the consequences of, an accident.

- General safety:
  - Confinement of radioactive materials should be provided by multiple zones.
  - The primary confinement zone should be the process ventilation system for spaces that may contain dispersible plutonium or other radioactive materials during normal operations.
  - A secondary confinement zone should be the operating and other potentially contaminated areas surrounding the process equipment.
  - A tertiary confinement zone should be provided in areas outside the secondary confinement zone to provide defense-in-depth between potentially contaminated areas and the environment.

- Pressure differentials should be maintained between building confinement zones and also between the building confinement zones and the outside atmosphere to ensure that airflow is from zones of lesser potential for contamination to zones of greater potential for contamination.
- Devices and alarms should be provided to indicate when pressure differentials are not maintained in a prescribed range.
- Ventilation systems may be designated as IROFS and require that the failure of any one component will not affect the continuous operation of the ventilation system.
- Ventilation systems designated as IROFS may need to be designed to withstand the effects of fires and to maintain confinement during these events.
- Components in ventilation systems designated as IROFS should be fire resistant to protect against fires that occur within or outside the system.
- Ventilation systems designated as IROFS should have fire and smoke suppression equipment to ensure that the integrity of high-efficiency particulate air (HEPA) filters or filter systems are not degraded during fires.
- Ventilation systems designated as IROFS should be designed to withstand tornado, hurricane, and other tropical storm conditions without the loss of confinement capability because of mechanical damage to the system or components or because of reduced ambient pressure at the intake and exhaust openings.
- Air monitoring and sampling systems should also be provided in occupied areas to alert workers of hazardous concentrations of radioactive or chemical materials.
- Gloveboxes designated as IROFS should be designed, fabricated, tested, installed, operated, and inspected in accordance with American Glovebox Society (AGS)-G001, “Guideline for Gloveboxes.”

12. RECOMMENDATIONS FOR REGULATORY GUIDANCE

The purpose of this section is to provide recommendations regarding guidance for designing off-gas and ventilation systems at spent fuel reprocessing facilities. The recommendations are based on the review of previous RGs pertaining to off-gas and ventilation systems (RG 3.20 and 3.32, respectively) and the review of the current state of aqueous reprocessing and pyrochemical reprocessing technologies, presented in this report. The assessment also considers the status of the regulatory framework pertaining to spent fuel reprocessing facilities, including the promulgation of the EPA regulation 40 CFR 190.

The classical PUREX process was designed to recover two product streams, plutonium oxide and uranium oxide, that are purified relative to fission products and minor actinides. Recent advances in aqueous reprocessing have focused on providing more options for chemical separations to meet the requirements of different fuel cycle applications and safeguard requirements. Examples include the co-recovery of uranium and plutonium together to avoid a purified plutonium product and the recoveries of selected minor actinides as a means of minimizing the radiological impact of the waste streams. These new developments have not fundamentally changed the separations chemistries with respect to fuel dissolution and chemical partitioning between aqueous and organic phases. Therefore, nothing in the existing RGs is necessarily obsolete or inadequate with respect to aqueous reprocessing and accompanying off-gas and ventilation system needs.

In contrast to aqueous reprocessing, pyrochemical reprocessing technologies are relatively new and still under development with respect to flowsheet unit operations, fuel cycle applications, safeguard requirements, and scaleup engineering. The existing RGs were developed at a time before pyrochemical reprocessing was under consideration for commercial scale reprocessing and were therefore not specifically addressed in the guidance. When the RGs were written in the mid-1970s, the Molten Salt Reactor Experiment (MSRE) at Oak Ridge National Laboratory (ORNL) had recently ended, and the IFR Program at Argonne National Laboratory (ANL) was 10 years away from beginning.

Off-gas and ventilation system requirements for pyrochemical reprocessing diverge from the requirements for aqueous reprocessing in a few noteworthy ways, as follows:[h]

1. Pyrochemical reprocessing does not use aqueous solutions and organic dilutants.
2. Pyrochemical reprocessing does not use concentrated nitric acids that generate nitrogen oxide off-gases and acid fumes during fuel dissolution.
3. Pyrochemical reprocessing uses high-temperature operations that create volatile fission product off-gases that would not necessarily be encountered during aqueous reprocessing. An exception is voloxidation that may be used in both aqueous and pyrochemical reprocessing.
4. Pyrochemical reprocessing uses molten chloride salts that capture a significant fraction of the fission product iodine but not so much as to preclude the need for iodine capture from the off-gas streams.
5. Pyrochemical reprocessing off-gases may be comprised mostly of argon from the argon atmosphere hot cells.
6. Pyrochemical reprocessing off-gases may contain more dry dust and condensable fumes.
7. Pyrochemical reprocessing off-gases present different challenges with respect to chemical compatibility and corrosion prevention. Examples include high-temperature process gases that contain oxygen, chlorine, and chloride salts.

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[h] There is no way of knowing exactly which aqueous and pyrochemical reprocessing flowsheets will be proposed for the construction of future licensed commercial reprocessing facilities. The descriptions and comparisons presented here are based on present understandings of these reprocessing technologies.

8. Pyrochemical reprocessing can potentially be used to reprocess younger spent fuel (i.e., fuel that has spent less time out of the reactor core prior to reprocessing). This is because the pyrochemical reprocessing reagents are not susceptible to radiation damage, as are in aqueous reprocessing reagents. Therefore, fuels are generally cooled for 5 years or more before aqueous reprocessing, while similar aging is not necessarily required for pyrochemical reprocessing. However, pyrochemical reprocessing must still manage the decay heat load from the buildup of fission products within the process equipment, which presents other challenges. The composition of radionuclides in the off-gases will be affected by fuel age.
9. Elements can behave differently in aqueous versus pyrochemical reprocessing. One example is iodine. In aqueous reprocessing, iodine can form compounds in both aqueous and organic solutions due to its complex valence chemistry. In pyrochemical reprocessing, it is thought that iodine accumulates in the salt, forming complexes with cesium. Iodine capture would need to be considered for both aqueous and pyrochemical reprocessing to meet 40 CFR 190, but there will likely be significant differences in iodine management techniques between the two reprocessing technologies.

Table 3 provides a more detailed comparison between aqueous and pyrochemical reprocessing off-gas considerations.

12.1. Functional Capabilities

A *functional capability* is a description of what the engineered system should do, while a *design constraint* is a description of how the engineered system is limited by factors that cannot be changed. For example, a functional capability statement is “the facility would have a capacity of no more than 50 units per day,” while a design constraint statement is “the site for the facility is limited to four acres of land.” Functional capabilities and design constraints do not specify how the engineered systems will do what they must do; instead they guide the designs of engineered systems toward common goals.

RGs 3.20 and 3.32 were written when some commercial reprocessing facilities were already operating or already under construction (see Appendix B). This explains why, in some instances, these early RGs provide very specific language for functional capabilities that may otherwise not be applicable in a general way to all foreseeable aqueous or pyrochemical reprocessing flowsheets.

The contents of RGs 3.20 and 3.32 can, however, be reduced to a few high-level functional capabilities listed below that cover the intent of the guides, which hinges on system reliability and safety. Ventilation and off-gas systems should:

1. Direct facility air flow from the areas of lower contamination potential toward the areas of higher contamination potential.
2. Maintain safe functionality during anticipated abnormal conditions such as process upsets, equipment malfunctions, losses of power, fires, severe weather, flooding, earthquakes, etc.
3. Be equipped with process monitors that alarm when abnormal conditions occur.
4. Provide means for process sampling.
5. Be constructed of materials that are heat-resistant and nonflammable and be equipped with smoke and fire suppression systems.
6. Have isolation devices that prevent the flow of contamination into uncontaminated areas during operations, inspections, maintenance, and abnormal conditions.
7. Designed such that components can be maintained, repaired, decontaminated, and replaced as needed.
8. Accommodate anticipated equipment and process changes.
9. Withstand the influx of materials such as process gases, dust, fumes, and condensates.

10. Withstand the deleterious effects of radiation and provide adequate radiation containment and shielding for worker safety.
11. Prevent the spread of contamination into unwanted areas and the environment.
12. Meet the standards of safety significant and safety class as identified by Federal guidance and regulations.

These functional capabilities may be considered when developing updated guidance later when the reprocessing technologies and designs being pursued for deployment are known with more certainty.

12.1.1. Other Resource Documents

Many documents have been written since the 1970s related to the best practices for the design, construction, operation, and risk assessment of nuclear facilities. Some of these documents contain information that is very relevant to the regulatory guidance for ventilation systems of nuclear fuel reprocessing facilities.

This report contains a section titled Technical Literature Bibliography, U.S. Government Documents, and the following several pertinent U.S. Department of Energy (DOE) documents are cited in this section, as follows:

- “Facility Safety,” U.S. Department of Energy, Order: DOE O 420.1C, December 2012
- “Nonreactor Nuclear Safety Design Guide for use with DOE O 420.1C, Facility Safety,” U.S. Department of Energy, Order: DOE G 420.1-1A, December 2012
- “DOE Standard: Development of Probabilistic Risk Assessment for Nuclear Safety Applications,” U.S. Department of Energy, DOE-STD-1628-2013, November 2013
- “DOE Handbook: Design Considerations,” U.S. Department of Energy, DOE-HDBK-1132-99, April 1999, Reaffirmed 2014
- “DOE Standard: Preparation of Nonreactor Nuclear Facility Documented Safety Analysis,” U.S. Department of Energy, DOE-STD-3009-2014, November 2014
- “DOE Standard: Natural Phenomena Hazards Analysis and Design Criteria for DOE Facilities,” U.S. Department of Energy, DOE-STD-1020-2016, December 2016
- “DOE Standard: Integration of Safety into the Design Process,” U.S. Department of Energy, DOE-STD-1189-2016, December 2016
- “DOE Standard: Air Cleaning Systems in DOE Nuclear Facilities,” U.S. Department of Energy, DOE-STD-1269-2022, April 2022
- “DOE Handbook: Handbook for Use with DOE-STD-1269-2022, Air Cleaning Systems in DOE Nuclear Facilities,” U.S. Department of Energy, DOE-HDBK-1169-2022, April 2022.

12.1.2. Recommendations

Existing RGs 3.18 and 3.20 continue to provide useful guidance with respect to their descriptions of functional requirements, in cases where these functional requirements are both high level and technology neutral. However, some of the functional requirements in these RGs are not useful because they are narrowly applicable to the reprocessing facilities that were licensed or pursuing licensing applications at that time. In other words, some portions of the RGs may not be applicable to the reprocessing technologies that are potential future candidates. It is recommended that designers and reviews consider the general high level functional capabilities noted in section 12.1., as well as the DOE documents listed above in section 12.1.1, to the extent they are relevant.

The development of updated guidance should be delayed until more is understood about how reprocessing will be pursued. Potential approaches to managing off-gases and volatile radionuclides are diverse and implementation scenarios will have wide variance based on the specific flowsheet designs. Specific capture requirements for each volatile radionuclide will be impacted by such choices as aqueous reprocessing versus pyrochemical reprocessing, flowsheet selection, facility design, and facility siting.

A future consideration is to consolidate the two RGs into a single guide that broadly contains the high-level functional capabilities listed above in addition to others that are yet to be determined. Specific functional capabilities bordering on engineering design requirements may be appropriate when design specifications for the next proposed reprocessing facility are known. However, at present, this is not the case. Design specifications for the next proposed reprocessing facility are unknown. There is also the future possibility of reprocessing advanced reactor fuel types that differ significantly from conventional LWR oxide and SFR metallic fuels. Therefore, consideration of overly specific and detailed functional capabilities should be avoided at present.

In many ways, the technology readiness of aqueous reprocessing is significantly higher than pyrochemical reprocessing. Nevertheless, significant advances have been made in both technologies since the prior RGs were written in the 1970s. Furthermore, since that time the DOE has published several orders, standards, and handbooks that contain guidance relevant to off-gas ventilation systems for reprocessing facilities. These and other references should be considered when developing new guidance.

13. DESCRIPTION OF VENTILATION SYSTEMS IN FCF AND HFEF

Spent fuel pyrochemical processing at INL occurs at two primary nuclear facilities at the Materials and Fuels Complex (MFC): the Fuel Conditioning Facility (FCF) and the Hot Fuel Examination Facility (HFEF). Exposed spent fuel is handled in inert-motivated hot cells to prevent undesired reactions with oxygen and moisture (i.e., water vapor). This section provides the design features and operational practices performed at these facilities to maintain the argon atmosphere and prevent releases of airborne contamination from the hot cells.

13.1. DOE Standards

DOE-STD-1269-2022, “Air Cleaning Systems in DOE Nuclear Facilities” provides requirements for safety class and safety significant air cleaning and confinement ventilation systems at DOE nuclear facilities and draws from applicable industry consensus codes where appropriate. New DOE nuclear facilities are built to this standard. The operations, testing, and maintenance provisions of the standard apply to existing facilities. Major modifications of existing facilities are required to adhere to the design requirements of DOE-STD-1269-2022 using the procedure in DOE-STD-1189-2016, “Integration of Safety into the Design Process.” DOE-HDBK-1169-2022, “Air Cleaning Systems in DOE Nuclear Facilities” contains best practices for facility design and accompanies DOE-STD-1269-2022.

Safety class[i] and safety significant[j] air cleaning systems are required to meet the requirements of DOE Orders DOE O 420.1C, “Facility Safety” and DOE O 420.1-1A, “Nonreactor Nuclear Safety Design Guide for use with DOE O 420.1C.” Both FCF and HFEF contain safety significant components that are relied upon to protect worker safety from radioactive hazards. Neither facility contains safety class components, which are necessary to mitigate hazards to the public.

DOE-STD-1269-2022 includes requirements for the following:

- The types of contaminants in the system
- Temperature control and ensuring the components of the system will function at the temperatures and environmental conditions to which they will be exposed
- Corrosive environments
- Emergency power
- Ensuring that pressure differentials in normal or upset conditions confine hazardous gases and particulates.

FCF and HFEF are legacy facilities that became operational in 1964 and 1970, respectively. The initial design and operation of both facilities greatly predates current revisions of DOE-STD-1269-2022 and DOE-HDBK-1169-2022.

Predecessor documents include ORNL-NSIC-13, “Filters, Sorbents, and Air Cleaning Systems as Engineered Safeguards in Nuclear Installations” issued in 1966, ORNL-NSIC-65, “Nuclear Air Cleaning Handbook: Design, Construction, and Testing of High-Efficiency Air Cleaning Systems for Nuclear Applications” issued in 1970, and ERDA-76-21, “Nuclear Air Cleaning Handbook: Design, Construction, and Testing of High-Efficiency Air Cleaning Systems for Nuclear Applications” issued in 1976. A crosswalk of period requirements and how they were met is unavailable for both FCF and HFEF. Within

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[i] *Safety-class structures*, systems, and components means the structures, systems, or components, including portions of process systems, whose preventive or mitigative function is necessary to limit radioactive hazardous material exposure to the public, as determined from safety analyses.

[j] *Safety-significant structures*, systems, and components means the structures, systems, and components that are not designated as safety-class structures, systems, and components, but whose preventive or mitigative function is a major contributor to defense-in-depth and/or worker safety as determined from safety analyses.

this document, the design and practices for argon atmosphere cleaning in both facilities are described, but a comprehensive verification matrix to contemporary requirements is unavailable.

13.2. HFEF Background

HFEF was constructed in 1970 to support post-irradiation examination research efforts. HFEF's mission includes examination and characterization of SNF from a variety of sources, research and development of radioactive waste forms, experimental scale pyrochemical processing of irradiated fuel, and storage of miscellaneous radioactive materials.

HFEF contains two adjacent shielded cells, an oxygen-atmosphere cell where operations for material handling take place, and an argon atmosphere main cell where operations with exposed spent fuel take place. The facility also contains a decontamination area and a repair area for hot cell equipment.

13.2.1. HFEF Argon Atmosphere System

The HFEF main cell is a negative pressure cell and designed to be leak-tight. It is a concrete-shielded, steel-lined enclosure with interior dimensions of 21.34-m long x 9.14-m wide x 7.62-m high and is filled with argon to provide an inert, non-oxidizing atmosphere designed to handle both confined and exposed radioactive materials. A summary of the design specifications of the argon atmosphere with respect to pressure, temperature, and atmosphere purity are given in Table 10. The engineered systems within the facility are designed to maintain the hot cell environment within these tolerances.

Table 10. Select design specifications of the HFEF argon atmosphere system.

	Normal	Minimum	Maximum
Cell Gauge Pressure	-249 to -995 Pa	-5474 Pa	+747 Pa
Cell Temperature	21 to 38°C	18°C	38°C
Cell Oxygen Content	60 ± 40 ppm	20 ppm	100 ppm
Cell Moisture Content	60 ± 40 ppm	20 ppm	100 ppm
	Normal Pressure	Minimum Recorded Pressure	Maximum Recorded Pressure
Site Barometric Pressure Conditions	84.86 kPa	82.15 kPa	87.00 kPa
	Maximum 24-hr change: 3386 Pa Maximum 1 hr. change: 339 Pa		
	Average	Summer	Winter
Mean Daily Pressure Change	508 Pa	339 Pa	677 Pa
Note: 1 atm = 101,325 Pa = 101.325 kPa			

The normal cell pressure range meets the DOE-STD-1269-2022 Section 3.2.2.12 that the primary confinement air pressure be at least 249 Pa lower than the secondary containment area[k] for process cells involving transuranic or alpha contamination.

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[k] The *secondary containment areas* in HFEF are the working floors surrounding the air-atmosphere and argon-atmosphere hot cells. The secondary containment areas in FCF are the working floors surrounding the argon-atmosphere hot cell and only a portion of the air-atmosphere hot cell where it connects to the argon-atmosphere hot cell. Many of the hot cell operations

The HFEF main-cell argon atmosphere system contains several subsystems:

- The main-cell cooling system
- The main-cell pressure and temperature control system
- Pressure limiting system
- Argon purification system.

13.2.1.1. Pressure and Temperature Control System

The main-cell pressure and temperature control system manipulates the argon- and cooling-fluid circulation systems, activating blowers, refrigeration units, closing and opening valves as needed to control main-cell pressure and temperature. Control panels are in a dedicated control room, and the operation of components can be done manually or automatically.

The system control compensates for issues such as heating from in-cell lighting, transfers into the main hot cell, and other operations that may affect the cell pressure such as maintenance on feedthroughs.

Main-cell cooling can be controlled to maintain either constant pressure or temperature. Pressure control is used most often. Pressure control minimizes the amount of cell gas that is vented to the atmosphere and the makeup argon that needs to be added back to the system. During these operations, the main cell is maintained at a pressure of -622 Pa (relative to barometric) and cell temperature between 21 and 32°C. An override control system ensures that the temperature does not exceed the maximum design temperature of 38°C for the main cell.

Temperature control is typically used for the following reasons:

- Constant temperature is needed for an experiment
- Operations require a larger than normal volume of argon to be discharged into the main cell, such as for the use of pneumatic tools
- Operations require a large volume of gas to be exhausted, which would result in temperatures near the upper temperature operational limits for an extended time.

13.2.1.2. Pressure Limiting Systems

Pressure limiting systems keep the cell pressure within specifications during events such as barometric pressure changes, loss of cooling, excessive cooling, excessive argon supply to the hot cell, and excessive argon removal from the cell. Some of these events may result from an auxiliary support system failing to engage when it should or erroneously engaging when it should not.

The main-cell normal argon supply provides argon from a cryogenic liquid argon storage tank located adjacent to the facility. Argon is fed into the cell from an argon header when the pressure falls below its setpoint, and this system is adequate to compensate for demand due to barometric pressure increases and normal argon usage.

The main-cell emergency argon supply provides argon from a manifold connected to 10 high-pressure gas cylinders. This system can quickly supply argon to the main cell if the cell pressure falls below emergency limits. The emergency argon supply is estimated to have enough argon for a day of normal usage, a 6.7°C decrease in cell temperature, or 60% of the maximum recorded barometric pressure increase for the area.

The normal exhaust system provides a backup to maintain negative pressure to the hot cell and operates when the cell is in the temperature control mode, a malfunction occurs, or to compensate for a

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staff work within the secondary containment areas. In other words, the insides of the hot cells have lower pressures than the secondary containment areas, and the secondary containment areas have lower pressures than the ambient atmosphere.

large barometric change. Argon diverted to the normal exhaust goes through a filter bank and is routed through an activated carbon prefilter to remove radioactive iodine, a prefilter to protect the HEPA filter from larger particulate, and a HEPA filter. An oil seal pot isolates the main-cell argon atmosphere from the cell exhaust system. The oil seal pot prevents back-diffusion of air into the cell if the cell exhaust valve is malfunctioning and fails to shut.

The overpressure and under-pressure relief system limits pressure deviations to prevent structural damage to the cell. When the overpressure setpoint is reached, a pressure relief valve enclosed in a seal pot automatically opens, and gas is exhausted through a separate stack on the outside of the building. Gas to be exhausted has already passed through the main-cell cooling system and has been filtered. When the pressure relief valve activates, a grab sample of the gas is automatically obtained. Air flows into the cell through the pressure relief valve and seal pot if the pressure reaches less than -5474 Pa (relative to barometric).

13.2.1.3. Main-Cell Cooling System

The main-cell cooling system includes an argon circulation system as shown in Figure 34. The argon circulation system circulates argon gas, filters particulate from the gas, cools the gas, and discharges it back into the main cell through overhead diffusers. The subcell is a space beneath the floor of the main cell. The subcell is a suspect radiation area and physical access to the subcell is controlled and used for access to mechanical systems when they require inspection or maintenance.

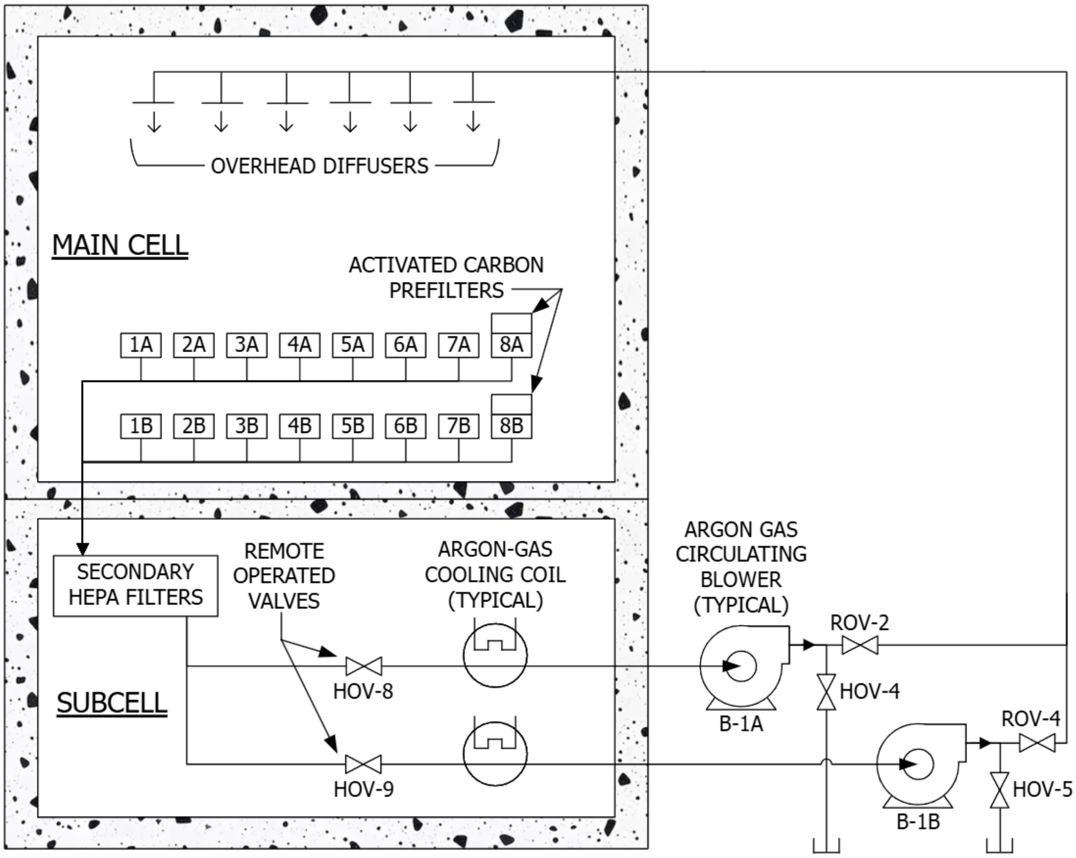

Figure 34. Schematic diagram of the HFEF argon circulating loop.

There are two separate cooling loops in the main cell, each controlled by a separate blower. The blowers are automatically controlled by the main-cell pressure and temperature control systems. During normal operation, at least one blower will normally be operating (lead blower), and if cooling demand increases, the second blower (lag blower) will automatically start. Either blower can be designated the lead blower. The system is designed such that failure of a single circuit will not reduce cooling system capacity enough to restrict operation in cell.

Two separate in-cell filter banks installed on the floor inside the hot cell remove particulate and radioactive iodine from the main-cell argon. Prefilters are positioned upstream from HEPA filters to collect coarse dust and particulate and prolong the life of the HEPA filters. Filter banks each contain eight HEPA filters/prefilters, seven of which recirculate directly to the cooling system. HEPA filters can be changed remotely by in-cell overhead handling equipment, and differential pressure is monitored across the filter to determine when changeout is needed.

The first HEPA filter on each filter bank has an activated carbon prefilter consisting of a tightly packed bed of absorbent carbon granule through which the argon gas is passed to remove radioiodine compounds, principally methyl iodide.

The headers from the main-cell filter banks join and penetrate downward through the main-cell floor into the subcell. Argon is routed to a secondary HEPA filter bank. In the event of a primary filter leak-through or spillage during filter/element replacement, secondary filters are designed to retain particulate and protect the system from excessive radioactive contamination.

Downstream of the secondary filters, the duct splits, and the argon gas flows through two parallel cooling coils, which are shell and tube heat exchangers with the argon to be cooled on the shell side and mineral spirits on the tube side. Each cooling coil uses a commercially manufactured refrigeration system to remove heat from the mineral spirits.

Downstream of the cooling coils, the argon ducts penetrate the subcell wall into a service-floor area where the blowers are located (one for each loop). Bearing temperatures of blowers are monitored so that their design temperatures are not exceeded. Argon from the two cooling coils is combined back into a single header. Diffusion headers discharge argon back into the main cell. The diffusers provide low velocity flow, which provides evenly distributed cooling and reduces the spread of contamination in the cell.

13.2.1.4. Argon Purification System

The argon purification system, and its connection to other facility systems, is shown in Figure 35. The argon purification system removes oxygen and moisture from the cell atmosphere to maintain the argon atmosphere of the cell. The purification system was designed to remove a maximum of 0.5% oxygen per pass. Nitrogen is not monitored in HFEF and can be removed from the cell atmosphere only by feed-and-bleed processes. Nitrogen is approximately 0.05% to 1% of cell atmosphere.

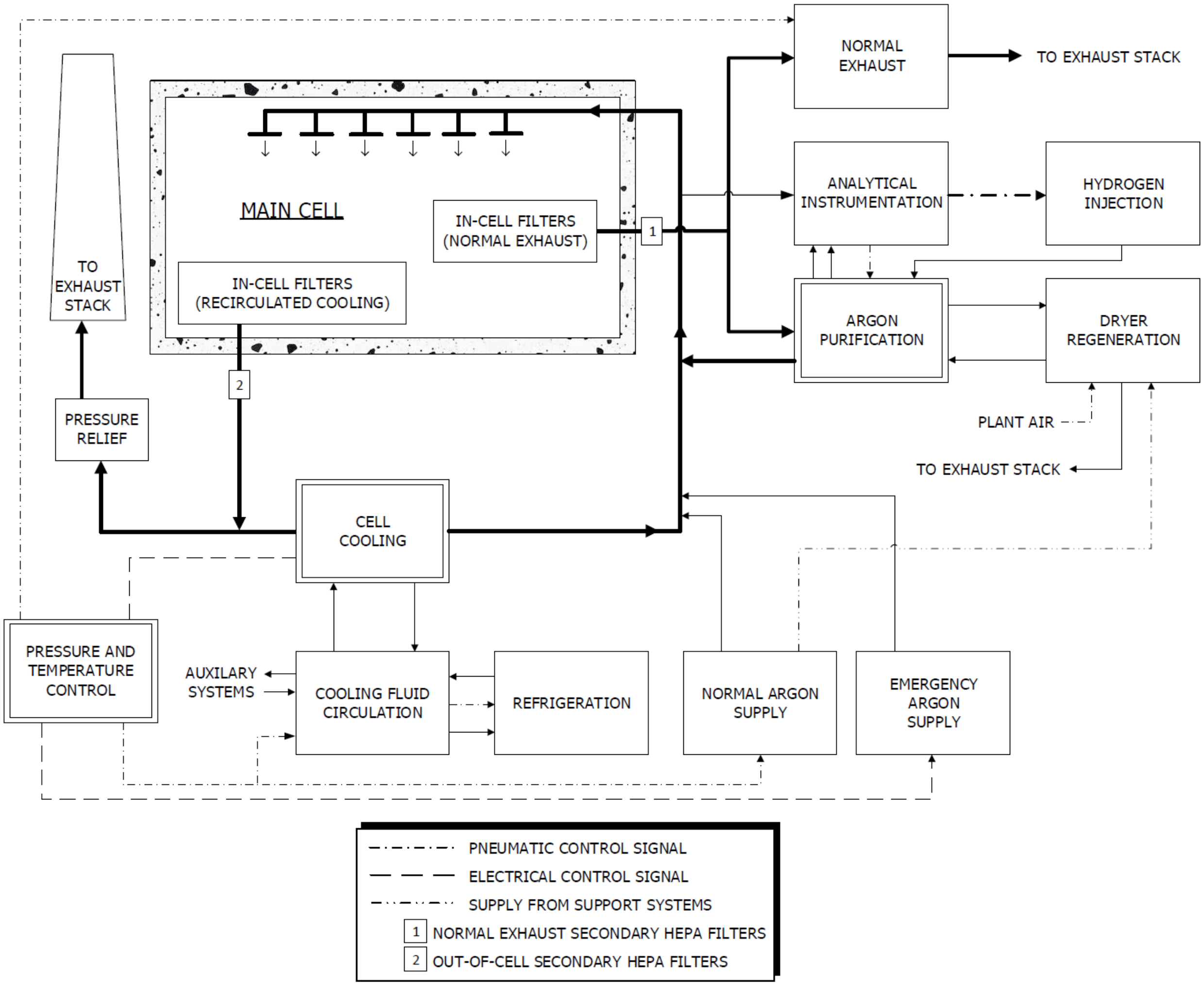

Figure 35. Schematic diagram of the HFEF argon cleaning system.

Gas to be purified is drawn from the normal exhaust duct rather than the main-cell cooling duct because the normal exhaust duct has 100% carbon filtering (in addition to HEPA filtering), whereas the cooling duct flow has only partial carbon filtering (in addition to HEPA filtering). The carbon filters eliminate gaseous contaminants that could poison the purifier catalyst. Catalyst poisons are listed in Table 11.

Table 11. HFEF catalyst poisons.

Contaminant	Warranty Limit (milligrams per cubic meter)
Sulfur	17,657
Phosphorous	17,657
Zinc	8,829
Collective Limit for P and Zn	17,657
Chromium	17,657
Nickel	17,657

Contaminant	Warranty Limit (milligrams per cubic meter)
Collective Limit for Cr and Ni	17,657
Silicon	35,315
Iron	176,574
Sodium	70,629
Calcium	70,629
Potassium	70,629
Collective Limit for Na, Ca, and K	70,629
Antimony	1,766
Arsenic	1,766
Copper	1,766
Lead	1,766
Tin	1,766
Lithium	1,766
Collective Limit for Sb, As, Cu, Pb, Sn, and Li	1,766
Mercury	177
Chlorine	883
Fluorine	883
Bromine	883
Iodine	883
Collective Limit for Cl, F, Br, and I	883

There are two parallel purification loops. In each loop, argon is forced from the normal exhaust via a blower into a purifier, where hydrogen is injected, and a palladium catalyst promotes the reaction between oxygen and hydrogen to form water vapor. Hydrogen injection is limited to a less than stoichiometric rate with the oxygen flow to limit hydrogen from being returned to the main cell. A shell and tube heat exchanger, with the argon gas on the shell side and mineral spirits on the tube side, cools the argon. Following gas cooling, the argon is directed through a dryer, which contains molecular-sieve material that absorbs moisture. An automatically operated valve can divert the flow around the dryer if moisture is less than 20 ppm; atmospheres with less than 20 ppm moisture create excess wear on in-cell components such as brushes in electrical motors.

Purified argon is diverted back to the main-cell cooling duct. Temperature and pressure sensing equipment is used to monitor and control the system and protect equipment (e.g., gaskets are protected against overheating). Detectors identify any leaks from the heat exchanger coolant.

When the molecular-sieve material in the dryer is saturated with water, it is regenerated by backflushing the molecular-sieve desiccant material with heated instrument air[1] and exhausting the air.

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[1] There are pneumatic process control instruments that operate on *instrument air*, which is a pressurized supply of clean dry air.

After regeneration, air is purged from the dryer with argon. Because the purification system contains two parallel loops, one dryer may be run to purify the argon while the other dryer is being regenerated. Under normal operations, each dryer is regenerated every 1.5 months.

Oxygen and moisture are continuously measured in the main-cell cooling duct to monitor main-cell gas conditions and at the discharge of both purification system dryers to assess purification effectiveness. Gas connections are available at these locations for calibration services of the oxygen and moisture sensors. Gas samples may be taken at these locations for laboratory analysis. A third gas sampling location is located at the discharge of both purification trains.

13.2.2. HFEF Cell Exhaust and Stack Monitoring

The cell exhaust system combines the normal exhaust from the HFEF argon cell with the exhaust streams from other areas within HFEF, including the HFEF air cell, radiological areas, gloveboxes, and pneumatic exhaust streams. Gas from higher contamination areas is passed through HEPA filters prior to entering the cell exhaust system. The combined exhaust stream is split into three parallel lines, each directing the exhaust through two HEPA filters in series, prior to being discharged from the exhaust stack.

As required by 40 CFR 61, “National Emission Standards for Hazardous Air Pollutants,” the HFEF stack monitor system continuously monitors alpha particulate, beta particulate, and beta gas. The system measures the airborne radioactive contamination content of the stack discharge and alarms if preset levels are exceeded. The system provides a digital record of activity levels exhausted from the main stack. Monitoring system equipment includes sampler assemblies, detectors, readout channels, a grab sample system, an interface module, a readout module, a pumping station for air collection, and system power source.

The sampling nozzles for this system are installed in the exhaust stack duct, where a continuous flow of sample is piped from the sample nozzles to the monitoring equipment and returned to the stack. Samples are split into three sample streams, with each stream initially passing through fixed-filter particulate samplers. Each stream goes through a different particulate sampler (i.e., alpha, gross beta, and ^{137}Cs). Gas in the alpha branch is fed through an iodine sampler with charcoal absorber cartridge. Gas from the other fixed-filter branches pass through samplers for gross beta and gamma (^{133}Xe and ^{85}Kr).

Components are required to be source checked weekly and after system calibration. The stack flowmeter is verified annually, and the sample flowmeter is calibrated quarterly. The filter cartridges in the fixed-filter samplers are collected manually and analyzed offline monthly. Additional components of the system require periodic maintenance, which is outlined in facility maintenance plans.

The system provides a means for collecting grab samples to be analyzed at an onsite analytical laboratory. A gas grab sample is taken automatically if the monitor alarms. Gas grab sampling can also be initiated manually.

13.2.3. HFEF Safety Significant Structures Systems and Components

Safety class structures systems and components (SSCs) are those for which credit must be taken, either preventive or mitigative, to meet the risk evaluation guidelines for the offsite public. There are no Safety Class SSCs in HFEF.

Safety significant SSCs are those for which the preventive or mitigative functions are major contributors to defense-in-depth (i.e., the prevention of uncontrolled radioactive or other hazardous material releases) and/or worker safety. HFEF main-cell passive components are considered safety significant. These include:

- Main-cell cooling system piping (including subcell HEPA filters)
- Main-cell normal exhaust system piping

- Main-cell normal exhaust ducts
- Main-cell emergency exhaust ducts
- Argon purification system piping
- Primary and secondary argon supply systems piping.

These components must be constructed of fire resistive and non-combustible materials and meet a leak rate criterion. Seal pots must maintain cell isolation from air ingress under normal and accident conditions.

The HFEF safety basis includes the following administrative requirements related to cell confinement, which require regular surveillance while the facility is in its high-material-at-risk operations mode:

- Main-cell passive components must be operable. An annual leak rate check is required for the entire argon cell confinement. Additionally, either a component or confinement boundary leak check is required following maintenance or modifications to confinement boundary.
- Seal pots on the main-cell normal exhaust system must be operable; their oil level is checked weekly when the cell pressure is greater than -995 Pa relative to the working floor.

The leak rate criteria for the main-cell confinement system passive components of 142 liters per minute is approximately 0.001% of main-cell volume and meets the maximum permissible leak rate standard for hot cells in DOE-STD-1269-2022, Section 3.2.2.12.

13.2.4. HFEF Standby Electrical Power

If normal electrical power for HFEF fails, the HFEF standby electrical power system activates. The system consists of two standby diesel generators and two uninterruptable power supplies (UPSs), which provide power to selected loads including the main-cell pressure and temperature controls, cell exhaust, radiation monitoring, stack effluent monitoring, and facility lighting.

Upon loss of normal power for greater than 3 seconds, both diesel generators start, and the lead diesel generator provides power to the selected loads via an automatic transfer switching system within 30 seconds of loss of normal power. Should the lead diesel generator fail, the second diesel generator assumes the electrical loads. The UPSs provide a constant supply of power for selected alarms, lighting, indications, and radiation monitoring equipment during periods of transition between normal and standby power.

HFEF standby power is considered neither safety significant nor safety class. In the event of loss of all electrical power, overpressure of the main cell can be prevented by venting through a manually operated valve and HEPA filters. If operator intervention is not possible, a passive seal pot and relief valve keeps the cell pressure from exceeding design limits and vents through HEPA filters. In the event of a loss of radiation monitoring to a specific area, the area is evacuated until monitoring can be returned to service. HFEF does not require continuous ventilation to prevent atmospheres that are an immediate hazard to life and health. Building ventilation can be deactivated for an extended period without resulting in a hazardous atmosphere. Stack effluent monitoring is not operational without normal or standby power. However, without operational building ventilation, stack effluent monitoring is unnecessary because there is no exhaust flow.

13.3. FCF Background

FCF was activated in 1963. Its original mission was demonstrating fuel processing and fabrication in a facility closely coupled to EBR-II. The mission was successfully carried out between 1964 and 1968. In the late 1960s, fuel processing equipment was removed, and FCF was refitted for examination of irradiation experiments from EBR-II, and until the mid-1970s, the facility provided destructive and nondestructive examinations of materials from EBR-II and the Transient Reactor Test Facility (TREAT). Throughout the late 1970s and the 1980s, FCF was used for repackaging of spent EBR-II fuel elements and continued with post-irradiation examination operations.

FCF's current mission centers on support to DOE Idaho Facilities Management Program (IFM) SFT Program. A large portion of the work is pyrochemical processing, including electrorefining, cathode processing, and casting development. The spent fuel used for these activities is primarily sodium-bonded metal fuels. Activities with oxide, mixed oxide, and nitride fuels may also be performed in FCF.

The major areas within the FCF facility include the following:

- An air atmosphere cell wherein fuel assemblies are disassembled into individual fuel elements
- An argon atmosphere cell wherein spent fuel elements are chopped and fuel conditioning operations take place
- A suited entry repair area where contaminated equipment can be cleaned and repaired
- An argon atmosphere glovebox where pyrochemical processing of unirradiated natural and DU is performed.

13.3.1. FCF Argon Atmosphere System

The FCF argon atmosphere hot cell is a circular room that encloses 1,700 cubic meters. The cell is bound by an operating corridor on the outside and a central observation room on the inside. Maintaining an argon atmosphere is important for both process and safety reasons due to the reactivity of the metals processed in the facility. The cell is maintained at a negative pressure with respect to the operating corridor, which is negative with respect to the outdoor ambient atmosphere. This prevents radiological release of contamination to the environment. Table 12 shows FCF argon cell operating and design characteristics.

Table 12. Select design specifications of the FCF argon atmosphere system.

	Normal	Minimum	Maximum
Cell pressure	-747 ± 50 Pa	-1493 Pa	-124 Pa
Cell temperature	10 to 41°C	10°C	41°C
Cell oxygen content	20 to 100 ppm	20 ppm	100 ppm
Cell moisture content	40 to 100 ppm	40 ppm	100 ppm
Cell nitrogen content	NA	NA	5,000 ppm

Pressures less than -1493 Pa cause increased leakage into the argon cell. A violation of the State of Idaho Air Quality Permit to Construct will occur if the argon cell pressure increases to the pressure of the air cell (approximately -124 Pa).

The FCF argon atmosphere system consists of three means of cell atmosphere control, as follows:

- The argon purification system
- The pressure and temperature control systems
- Cell feed-and-bleed system.

For facility documentation purposes, the HFEF air cell exhaust system is not considered part of the FCF atmospheric control system, but it interfaces with both the argon feed-and-bleed system and the argon purification system. See Figure 36 for a simplified diagram of the argon flow pathway through FCF.

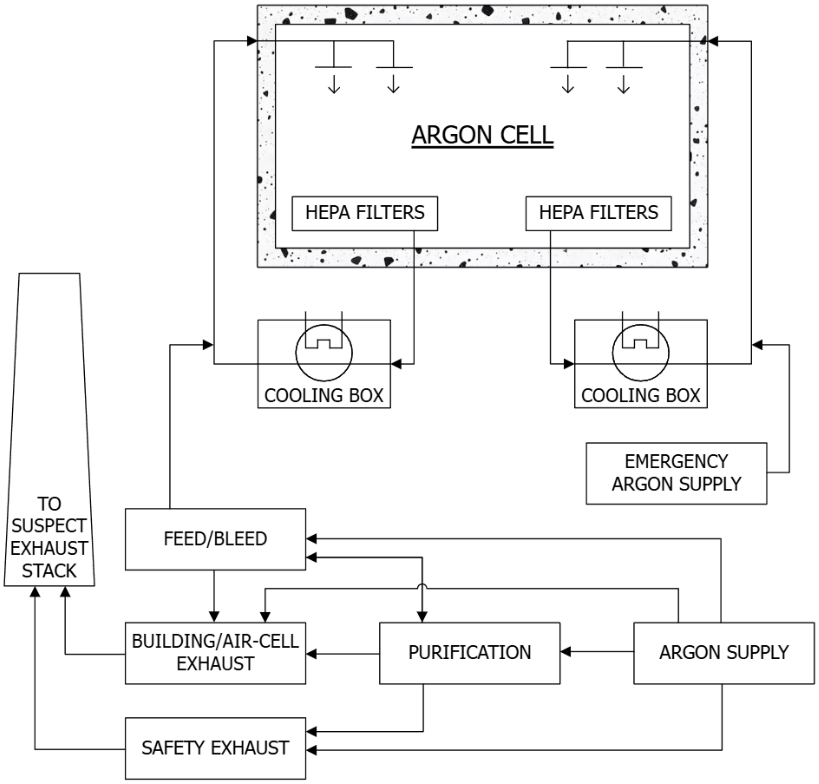

Figure 36. Schematic diagram of the FCF argon flow pathways.

13.3.1.1. Pressure/Temperature Control System

The FCF pressure and temperature control system shares many similarities with the HFEF pressure and temperature control system. There are multiple operator control stations. Redundant processors in combination with the process logic controllers automatically manipulate cooling and argon valves to control argon temperature and pressure. The system may also be controlled manually. Like the HFEF system, the FCF system may be operated in pressure or temperature control mode. The normal pressure control setpoint is -747 Pa with a normal pressure range of -697 to -796 Pa. The normal argon cell temperature is 10 to 41°C. When in the temperature control mode, the temperature is controlled using the temperature sensed at the inlet of one of the two recirculation loops.

13.3.1.2. Argon Recirculation and Cooling

Like the cooling system in HFEF, the argon in FCF is drawn from the cell through HEPA filters, cooled, and then discharged back into the FCF argon cell (Figure 36). There are two separate recirculation loops in FCF. The loops can either be operated in parallel or independently, allowing for alternate shutdown of loops for maintenance. Each loop has its own blower. The south recirculation loop connects to the cell purification system, which will be discussed later.

Argon gas is drawn into each recirculation loop through HEPA filter banks located in the argon atmosphere cell. The northside recirculation filter bank contains 8 HEPA filters, and the southside recirculation loop has 12 HEPA filters. Prefilters are located at the inlet to the north loop to extend HEPA filter life. Filters may be replaced remotely. Unlike HFEF, FCF does not have activated carbon scrubbers in the cooling loop to remove iodine.

Following filtration, argon from each loop is drawn into separate cooling boxes each located within the FCF subcell. Each cooling box houses four evaporator coils to cool the argon. The flow of the refrigerant (R-22, chlorodifluoromethane) is controlled by the pressure and temperature control systems.

Each cooling loop is equipped with a freon detection system that monitors the presence of refrigerant in the argon stream. Facility cooling water is used to remove heat from the refrigerant.

Blowers (one for each loop) are located downstream of the cooling boxes. The fan housing is equipped to withstand full vacuum. Blowers are equipped with temperature indicators to monitor bearing temperatures. Cooled argon is discharged into the FCF argon cell downstream of the blowers.

13.3.1.3. Argon Purification System

The FCF argon purification system continuously monitors the argon cell atmosphere for oxygen, nitrogen, and moisture. The purification system loop is in the FCF subcell. Argon is fed into the purification system downstream of the south loop argon cooling box and returned to the south cooling loop upstream of the cooling box. Like the HFEF purification system, the FCF argon purification system removes oxygen by hydrogen injection in the presence of a catalyst. Moisture is removed by a desiccant bed. The purification system consists of a blower, two parallel catalyzers, two parallel dryers, a dryer regenerator, and analytical instrumentation. Impurity limits for fuel processing quality control are 20 ppm to 100 ppm for oxygen, 40 to 100 ppm for water vapor, and <5,000 ppm for nitrogen. Nitrogen can only be removed from the hot cell atmosphere via feed-and-bleed process.

Argon from the south cooling loop is first passed through a HEPA filter followed by the blower. Hydrogen is injected downstream of the blower prior to the catalyzers. There are two catalyzers that can be configured in parallel, series, or both bypassed depending on the valve lineup.

The catalyzers are cylindrical vessels that contain 9.525-mm diameter alumina spheres loaded on top of the catalyst. The catalyst (DeOxo Catalyst, Type A) consists of palladium-bonded 3.175-mm diameter alumina pellets. The catalyst promotes the reaction between hydrogen and oxygen to form water vapor. Each catalyzer can remove up to 1% of oxygen from an argon gas stream flowing at over 11,000 liters per minute.

Following the catalyzers the argon flow is directed through parallel dryers, and flow can also be diverted around the dryers. The absorptive desiccant (Linde Division Type 5A Zeolite) in each dryer consists of 3.175-mm diameter molecular-sieve, alumina-silica pellets. Following drying, the argon is returned to the south cooling loop upstream of the cooling box.

Dryers are regenerated with heated dry plant air and then purged with argon. Exhaust from regeneration operations flows through the top of the dryer and through the air cell exhaust system. Due to the parallel configuration of the dryers, one dryer may be regenerated while the other dryer is operational.

Instrumentation to monitor the purification system includes flow indicators, temperature indicators, pressure gauges, nitrogen analyzers, oxygen analyzers, moisture analyzers, and hydrogen leak detectors. The gas analyzers are periodically calibrated. Cylinders of 1.6 vol% nitrogen in argon, 50–200 ppm oxygen in argon, and 4.0 vol% hydrogen in argon are used for calibration purposes.

13.3.1.4. Argon Feed-and-Bleed System

The argon feed-and-bleed system provides clean argon to the cell and process equipment, such as electrorefiners, the cathode processor, and the casting furnace. The system also removes excess argon to equalize pressure or decrease nitrogen concentration. Nitrogen is considered an impurity in the argon atmosphere and can be introduced into the cell through in-leakage through seal pots or from transferring items into or out of the cell. Addition of fresh argon into the cell, and purging of existing argon, dilutes the nitrogen concentration to desired levels. The feed-and-bleed system uses the nitrogen concentration measured by the purification system to signal when argon needs to be fed or purged.

The feed-and-bleed system works in concert with the pressure and temperature control systems to maintain argon cell pressure, temperature, and concentrations of oxygen, moisture, and nitrogen within specified parameters.

The feed-and-bleed system consists of three subsystems, as follows:

- An argon feed system supplies argon gas to the argon cell and process equipment. An outdoor dewar feeds argon to the FCF casting furnace as well as to a surge tank. The surge tank supplies the argon cell and other system needs and acts as a reservoir to minimize pressure and flow effects of sudden system demands.
- An argon bleed system removes argon from the cell. Gas is drawn from the argon cell purification line, and off-gas is directed to the air cell exhaust system and out of the stack to atmosphere. Prior to discharge into the air cell exhaust system, the argon passes through a HEPA filter, two carbon adsorbers, and a post filter to remove particulate and radioactive gases. The two carbon adsorbers are used in series to meet the residence time for iodine removal. The HEPA filter is changed biennially and the carbon adsorber is changed annually.
- An emergency argon supply system quickly provides argon to the hot cell if needed. This consists of 10 high-pressure cylinders of argon gas that automatically release into the cell via the argon recirculation system to increase falling cell pressure. The system actuates if the argon cell pressure reaches a predetermined setpoint.

13.3.2. FCF Cell Exhaust and Stack Monitoring

The safety exhaust system actively filters discharge of particulates from the argon cell and selected subcell areas during and after a postulated design basis accident. The system automatically activates when the cell pressure reaches -248 Pa, which is the minimum pressure differential of the primary confinement for a hot cell as required by DOE-STD-1269-2022, Section 3.2.2.12. The system contains two identical independent “trains.” If one train fails to operate, the second train automatically activates. Each train contains one branch to exhaust the argon cell and another to exhaust subcell areas. The two branches pass through HEPA filters. Each argon branch contains a seal pot to prevent back mixing of air into the argon branch and provide under-pressure and overpressure protection to the argon cell structure in the event of pressure anomalies. The air and argon branches are combined and drawn through another set of HEPA filters before they are sent to the exhaust stack. A simplified diagram of the system is shown in Figure 37.

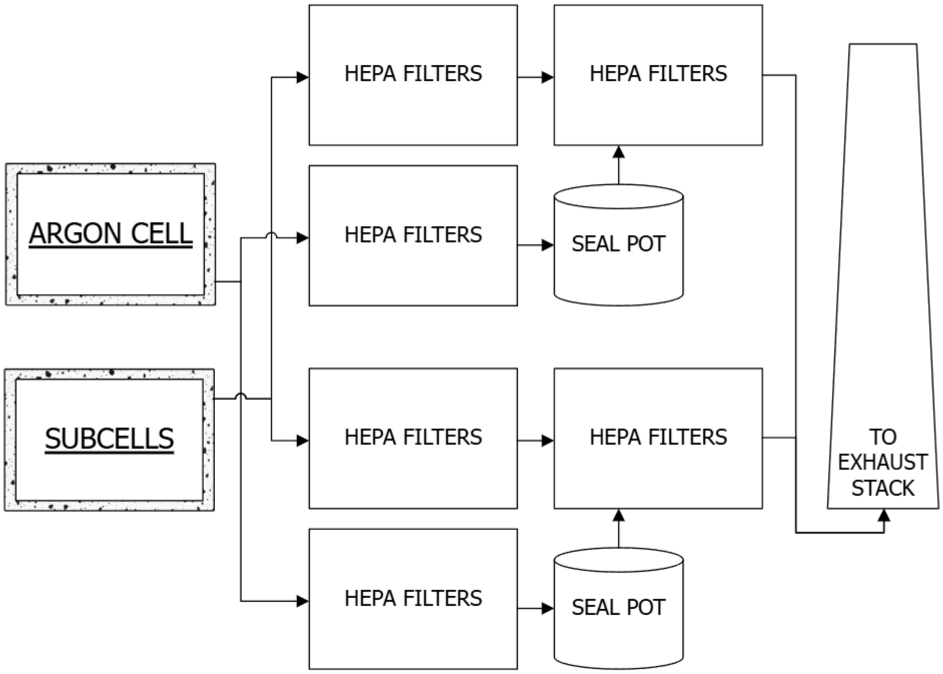

Figure 37. Schematic diagram of the FCF exhaust pathways.

The FCF stack monitoring system monitors and records airborne releases from the FCF exhaust stack as required by 40 CFR 61, "National Emission Standards for Hazardous Air Pollutants," which requires that "all radionuclides which could contribute greater than 10% of the potential effective dose equivalent for a release point shall be measured." Calculations have been made to determine those radionuclides for FCF. The following radionuclides have been selected for monitoring either by on-line or by offline sample analysis methods: ^{90}Sr/^{90}Y, ^{137}Cs/^{137m}Ba, and ^{239}Pu. The list of radionuclides to be monitored may be updated as operational experience increases and facility processes change.

Gas to be monitored is withdrawn from a vertical portion of the FCF stack at an elevation of approximately 17.4 m. This sampling point is 10 diameters downstream of the last major flow direction change and meets the requirement of a minimum of 5 diameters specified in ANSI N 13.1-l969. The system is composed of two parallel subsystems that are separate but functionally identical. The stack is sampled at eight withdrawal points for each subsystem (a minimum of six are required by ANSI N13.1-1969) per probe, with one probe per stack monitor subsystem.

The sample stream in the stack is piped from the withdrawal point to the monitoring equipment in the facility. Each subsystem has identical real-time radiation monitoring for alpha particulate (^{239}Pu), beta particulate (^{137}Cs and ^{90}Sr), and noble gas (^{85} Kr). Alarms and alerts are transmitted to the FCF control room based on adjustable limits. After the gas passes through the detectors, it is returned to the stack. Monitoring equipment is calibrated every 6 months and source checked weekly. Filters from particulate monitors are regularly removed and analyzed to determine the concentrations of radionuclides required for monitoring by 40 CFR 61.93, "Emission Monitoring and Test Procedures." These filters are typically analyzed monthly. The system can collect gas grab samples from each stack monitor subsystem. Gas sample containers can also be filled when a signal is initiated manually. Counting two different samples collected at different times can help determine the nature of any release should an alarm setpoint be reached on the continuous monitoring equipment.

13.3.3. FCF Safety Significant Structures Systems and Components

Like in HFEF, there are no safety class SSCs in FCF.

The FCF argon cell confinement boundary is considered safety significant and includes the following:

- Passive penetrations (penetrations not normally opened) into the argon cell
- Recirculation, purification, cooling, and ductwork
- Safety exhaust system ductwork and seal pots.

The safety functions of the confinement boundary are to confine radioactive materials within the argon cell and prevent air ingress to the argon cell, which precludes the possibility of pyrophoric fires.

The integrity of the argon cell confinement boundary is designed, constructed, and tested to perform as a barrier to gas and airborne radioactive and hazardous material transmission between the argon cell and surrounding areas. Any changes to this boundary must be configuration managed.

The FCF safety basis includes the following administrative requirements related to cell confinement, which require regular surveillance while the facility is in operations mode, as follows:

- Argon cell passive components must be operable. An annual leak rate check is required for the entire argon cell confinement. Additionally, a leak rate check is required following maintenance or modifications to safety significant components of the confinement boundary with openings greater than 6.35-mm equivalent diameter.
- Seal pots in the safety exhaust system must be operable; the oil level within them is checked weekly when the argon cell pressure is greater than or equal to -995 Pa.

The leak rate criteria for the FCF argon cell confinement system passive components of 142 liters per minute is approximately 0.001% of main-cell volume and meets the maximum permissible leak rate standard for hot cells in DOE-STD-1269-2022, Section 3.2.2.12.

13.3.4. FCF Standby Electrical Power

Like HFEF, FCF standby electrical power consists of diesel generators and UPSs to power similar systems as HFEF standby electrical power including exhaust systems, exhaust stack monitoring, lighting, and hot cell atmosphere control systems. Also, like the HFEF system, FCF standby power is considered neither safety significant nor safety class.

13.4. Summary

An overview of argon cleaning systems in the HFEF and FCF hot cells was described based on available documentation. The systems used in these facilities are similar but include small differences in how argon is cooled, purified, cleaned, filtered, monitored, and vented. Both facilities contain redundant systems to keep argon cell temperature, pressure, and argon purity within specifications and prevent radiological release to the environment.

14. TECHNICAL LITERATURE BIBLIOGRAPHY

This section provides a bibliography of select technical literature found during the preparation of this report. A wide net was cast in the collection of citations, but all are relevant to the subject of nuclear fuel reprocessing. The citations are arranged in approximate chronological order within each of the following five categories:

- U.S. Government Documents

  This collection includes documentation from government agencies such as the U.S. Atomic Energy Commission, U.S. EPA, U.S. Department of Energy, and U.S. Nuclear Regulatory Commission.

- Air Cleaning Conference Proceedings

  This conference series began in 1951 and occurred on a mostly biennial basis up to 2024. The conference proceedings are a good source of information on subjects related to nuclear fuel reprocessing.

- Scientific Journal Literature

  There are some examples of redundancy between the citations in Scientific Journal Literature and citations in National Laboratory and Corporate Reports. This most likely occurs when a report serves as the basis for a manuscript.

- National Laboratory and Corporate Reports

  These citations are limited to documents available to the public. Some of these citations started as classified information and were later declassified.

- International Atomic Energy Agency (IAEA) Reports

  These citations have an international perspective. Presently, only France and Russia are actively reprocessing nuclear fuels at a commercial industrial scale.

- International Organization of Standards (ISO)

  This organization develops standards for a broad spectrum of industrial sectors.

- American Society of Mechanical Engineers (ASME)

  This organization develops standards for mechanical systems.

- American Glovebox Society

  This organization develops standards for nuclear gloveboxes.

14.1. U.S. Government Documents

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14.2. Air Cleaning Conferences Proceedings

These conferences were sponsored by the DOE and NRC up to 26th, and by the International Society of Air Treatment Technologies from 27th onward. The complete series of conferences is listed in Table 13.

Table 13. Chronology of Clean Air Conferences

Conference Number	Date	Site	Document Number	Number of Volumes
1	June 12–15, 1951	Boston, MA	No Report	
2	Sept. 15–17, 1952	Ames, IA	WASH-149	1
3	Sept. 21–23, 1953	Los Alamos, NM	WASH-170	1
4	Nov. 2–4, 1955	Argonne, IL	TID-7513(Pt. 1)	1
5	June 24–27, 1957	Boston, MA	TID-7551	1
6	July 7–9, 1959	Idaho Falls, ID	TID-7593	1
7	Oct. 10–12, 1961	Upton, NY	TID-7627	1
8	Oct. 22–25, 1963	Oak Ridge, TN	TID-7677	1
9	Sept. 13–16, 1966	Boston, MA	AEC-660904	2
10	Aug. 28, 1968	New York, NY	CONF-680821	1
11	Aug. 31–Sept. 3, 1970	Richland, WA	CONF-700816	2
12	Aug. 28–31, 1972	Oak Ridge, TN	CONF-720823	2
13	Aug. 12–15, 1974	San Francisco, CA	CONF-740807	2
14	Aug. 2–4, 1976	Sun Valley, ID	CONF-760822	2
15	Aug. 7–10, 1978	Boston, MA	CONF-780819	2
16	Oct. 20–23, 1980	San Diego, CA	CONF-801038	2
17	Aug. 2–5, 1982	Denver, CO	CONF-820833	2
18	Aug. 12–16, 1984	Baltimore, MD	CONF-840806	2
19	Aug. 18–21, 1986	Seattle, WA	NUREG/CP-0086 CONF-860820	2
20	Aug. 22–25, 1988	Boston, MA	NUREG/CP-0098 CONF-880822	3

Conference Number	Date	Site	Document Number	Number of Volumes
21	Aug. 13–16, 1990	San Diego, CA	NUREG/CP-0116 CONF-900809	2
22	Aug. 24–27, 1992	Denver, CO	NUREG/CP-0130 CONF-9020823	2
23	July 25–28, 1994	Buffalo, NY	NUREG/CP-0141 CONF-940738	1
24	July 15–18, 1996	Portland, OR		
25	Aug. 3–6, 1998	Minneapolis, MN		
26	Sept. 11–12, 2000	Richland, WA		
27	Sept. 23–25, 2002	Nashville, TN		
28	Sept. 27–29, 2004	Albuquerque, NM		
29	July 19–21, 2006	Cincinnati, OH		
30	Aug. 25–27, 2008	Seattle, WA		
31	July 19–21, 2010	Charlotte, NC		
32	June 17–19, 2012	Denver, CO		
33	June 22–24, 2014	St. Louis, MO		
34	June 5–7, 2016	San Antonio, TX		
35	June 3–5, 2018	Charleston, SC		
36	June 28–29, 2022	Salt Lake City, UT		
37	Sept. 8–10, 2024	Knoxville, TN		

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  - P. D. Gildea, H. G. Birnbaum, W. R. Wall, “Modification and Testing of the Sandia Laboratories, Livermore Tritium Decontamination Systems”
  - F. B. Hill, Y. W. Wong, Y. N. Chan, “Tritium Removal Using Vanadium Hydride”
  - K. Hata, Y. Nishizawa, Y. Osawa, “Study on the Tritium Removal from the Sodium in LMFBR”
  - M. J. Kabat, “Monitoring and Removal of Gaseous Carbon-14 Species”

- R. D. Penzhorn, K. Gunther, P. Schuster, "On the Catalytic Removal of Ozone Produced in Radioactive Media"
- Session 5 Adsorbers and Absorbents
- R. R. Bellamy, V. R. Deitz, "Confirmatory Research Program – Effects of Atmospheric Contaminants on Commercial Charcoals"
- J. B. Romans, V. R. Deitz, "A Non-Radioactive Determination of the Penetration of Methyl Iodine Through Impregnated Charcoals"
- A. J. Juhola, J. V. Friel, "Effect of Pore Structure on the Activated Carbon's Capability to Sorb Airborne Methylradioiodide"
- G. O. Wood, C. A. Kasunic, "Methyl Iodine Retention on Charcoal Sorbents at Part-Per-Million Concentrations"
- J. L. Kovach, "Evaluation and Control of Poisoning of Impregnated Carbons Used for Organic Iodine Removal"
- V. R. Deitz, "A Determination of the Attrition Resistance of Granular Charcoals"
- T. R. Thomas, B. A. Staples, L. P. Murphy, "The Development of Ag°Z for Bulk 120I Removal from Nuclear Fuel Reprocessing Plants and PbX for 120I Storage"
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- "D. T. Pence, C. C. Chou, J. D. Christian, W. J. Paplawsky, "Noble Gas Separation with the Use of Inorganic Adsorbents"
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- J. A. Stone, D. R. Johnson, "Measure of Radioactive Gaseous Effluents from Voloxidation and Dissolution of Spent Nuclear Fuel"
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- M. N. Elliot, E. Lilleyman, "A Review of Some U. K. A. E. A. Work on Gas Cleaning in Fuel Reprocessing Plants"
- A. Bruggeman, L. Meynedonckx, W. R. A. Goossens, "Elimination of Nox by Selective Reduction with NH3"
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14.6. International Organization of Standards (ISO)

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- ISO 10648-1:1997(en): Containment enclosures –Part 1: Design principles
- ISO 11933-2:1997(en): Components for containment enclosures –Part 2: Gloves, welded bags, gaiters for remote–handling tongs and for manipulators
- ISO 11933-3:1997(en): Components for containment enclosures –Part 3: Transfer systems such as plain doors, airlock chambers, double-door transfer systems, leak-tight connections for waste drums.
- ISO 17873:2004(en): Nuclear facilities–Criteria for the design and operation of ventilation systems for nuclear installations other than nuclear reactors
- ISO 15080:2001(en): Nuclear facilities –Ventilation penetrations for shielded enclosures

14.7. American Society of Mechanical Engineers

- “In-Service Testing of Nuclear Air-Treatment, Heating, Ventilating, and Air-Conditioning Systems,” The American Society of Mechanical Engineers, ASME N511-2022
- “Code on Nuclear Air and Gas Treatment,” The American Society of Mechanical Engineers, ASME AG-1-2023

14.8. American Glovebox Society

- “Guidelines for Gloveboxes, Third Edition,” American Glovebox Society, Standards Development Committee, AGS-G001, February 2007

Appendix A

This appendix contains copies of NRC RGs 3.18, 3.20, and 3.32, which are related to off-gas and ventilation systems for reprocessing facilities. The official versions of these guides can be found in Agencywide Documents Access and Management System (ADAMS), NRC's official recordkeeping system.

A-1. NRC Regulatory Guide 3.18

CONFINEMENT BARRIERS AND SYSTEMS FOR FUEL REPROCESSING PLANTS

A. INTRODUCTION

Section 50.34 of 10 CFR Part 50, "Licensing of Production and Utilization Facilities," requires, among other things, that each application for a construction permit for a production or utilization facility, including a fuel reprocessing plant, include the principal design criteria for the facility. Section 20.1 of 10 CFR Part 20, "Standards for Protection Against Radiation," states that licensees should make every reasonable effort to maintain radiation exposure, and releases of radioactive materials in effluents to unrestricted areas as far below the limits specified in that part as practicable. Properly designed confinement, barriers and systems in fuel reprocessing plants provide a principal means of reducing such exposures and releases. This regulatory guide provides information relative to establishing principal design criteria for confinement systems that will minimize the amount of radioactive material released to the environment or to areas normally occupied by personnel.

B. DISCUSSION

A principal objective in the design of fuel reprocessing plants is to prevent the uncontrolled release and dispersal of radioactive materials. These materials are ingredients of process fluids, process solids, and building ventilation gases. Release of radioactive materials is controlled by one or more individual confinement barriers and systems which successively restrict releases of radioactive material to the environment or into areas normally occupied by plant personnel. The confinement system design is affected by the design and operation of building ventilation and process off-gas systems and by fire protection requirements. These items will be subjects of future guides.

Fuel cladding, fuel canisters, product or waste storage containers, or walls of process vessels or piping commonly provide the first confinement barrier to control releases of radioactive material. Additional barriers, as necessary for safety, are provided by ancillary process systems and equipment such as the process off-gas system or gloveboxes, by process cell or vault walls, or by building walls. These additional barriers are designed to work in conjunction with the building ventilation system to limit the spread of airborne contamination which could be generated by leakage of either gases or liquids from process and storage vessels into process cell or vault air spaces and thence into other areas. Barriers also serve to prevent the spread of contamination via leakage of contaminated liquids and solids.

In order to limit the spread of radioactive materials, processing facilities are separated by confinement barriers into areas or zones of various levels of contamination. The building ventilation and process off-gas systems aid in controlling the spread of airborne contamination through openings in barriers by regulating the direction of air or gas flow between these zones so that gas leakage is successively from zones of low potential for contamination to zones of higher potential for contamination. For example, air flow would be successively from the environs to building areas occupied by personnel, to potentially contaminated process areas, to the ventilation cleanup system, and thence to blowers for discharge to the atmosphere. The capacity of ventilation systems must be adequate in relation to confinement system requirements to ensure that the velocity of gas flowing through any barrier opening is sufficient to prevent backflow of airborne contaminants through such openings and that air flow patterns are not disrupted by winds, movement of equipment or personnel, or temporary opening of passageways through confinement barriers.

Heat transfer fluids such as water, steam, and air provide potential pathways for release of radioactive materials to the environment. These fluids often circulate between radioactive and nonradioactive areas and are subject to contamination by leakage of process solutions through defects which may develop in heat exchanger components. Should these fluids be discharged to the environs, releases of radioactivity may occur. Barriers to release via this pathway are usually provided by intermediate heat exchangers located between the utility system and the process system. This serves to form a “closed-loop” (as opposed to a “once-through”) heat transfer circuit in which the heat exchangers function as confinement barriers.

Piping penetrations through confinement barriers provide another potential pathway for release of radioactive material to the environment. These penetrations usually serve as conduits for instrument leads or serve to conduct cold chemical or utility fluids between zones formed by the barriers. During maintenance or remodeling activities, it may be necessary to isolate selected equipment items to assure confinement. In such penetrating lines an isolation valve located close to the confinement barrier can function as a confinement barrier by being closed during such activities.

In this guide, barriers are arbitrarily classified as “total” or “selective.” Total barriers are those fabricated of impermeable materials which can be expected to prevent penetration of all confined material without regard to that material’s physical or chemical nature. Examples of total barriers are pipe and vessel walls and walls of buildings and structures. Total barriers may also function as radiation shields. Information on design for this function may be found in Regulatory Guide 3.9. Selective barriers are mass transfer devices or filters and are usually employed to remove selected chemicals or particulate matter from a process or ventilation stream while allowing the bulk amount of the stream to pass through. Examples of selective barriers are adsorbers, scrubbers, distillation units, and particulate filters.

To verify continued integrity of barriers, monitoring devices are necessary. Where a total barrier is used for gas confinement, integrity may reasonably be inferred by monitoring the difference in pressure across the barrier and noting any deviation from normal. Where a total barrier is used for liquid confinement, leakage may be indicated by a liquid high-level alarm in a specially designated leakage collection sump or by analytical devices which measure and alarm contaminant intrusion into normally uncontaminated tanks or process streams. Continuous air monitors are used to indicate any breach in integrity of a total barrier or any loss of function of a selective barrier which would release radioactivity from a designated confinement area into a monitored area.

C. REGULATORY POSITION

1. Confinement systems and barriers should be provided to limit releases of radioactive material to restricted areas so that exposures to individuals are as low as practicable and that the concentrations are as low as practicable and do not exceed the limits specified in §20.103 of 10 CFR Part 20.
2. Confinement systems and barriers should be provided to limit releases of radioactive materials to unrestricted areas; they should be capable of withstanding the effects of design basis accidents, including natural phenomena, so that releases of radioactive materials are as low as practicable and so that they do not exceed the annual limit prescribed by §20.106(a) of 10 CFR Part 20.
3. Pipes penetrating confinement barriers should be equipped with at least one isolation valve located outside of and as close to the confinement barrier as is practicable.
4. Confinement zones should be established and should be bounded by confinement barriers. Generally, zones should be established on the basis of their function(s) which may include confinement of process fluids, process equipment, maintenance areas, operating areas, and process control areas.

Zones should be arranged in order of contamination potential. The zones with the highest contamination potential should be situated adjacent to contaminated process areas and the zones with the lowest contamination potential at the outer areas of the process building.

5. Barriers should be designed with regard to the capabilities of the process off-gas and the building ventilation systems. Barriers should be designed to withstand loadings due to pressure differentials imposed by these systems. Openings through barriers should be limited in size to preclude disruption of confinement or disruption of air flow patterns due to loss of pressure differential.
6. Barriers should be provided to prevent leakage of contaminated fluids into heat transfer fluids circulated into unrestricted areas. A preferred method for this is by use of closed primary and independent secondary heat transfer circuits.
7. To monitor the integrity of total confinement barriers for gases, devices should be provided to control and indicate pressure differentials between confinement zones. Alarms should be provided to indicate when pressure differentials are not maintained within a prescribed range. In addition, radiation monitoring devices with local alarms should be provided to indicate leakage of radioactive material through confinement barriers into normally inhabited areas.
8. To monitor the integrity of total confinement barriers for liquids, devices should be provided to indicate and alarm upon liquid leakage through a confinement barrier.
9. To monitor the function of selective barriers, devices should be provided to indicate and alarm any deterioration or loss of function which may release radioactivity from designated confinement zones into other zones or to the environs. Devices such as CAMs (continuous air monitors) and stack monitors are acceptable.
10. Confinement systems should be constructed of nonflammable materials except where this is not possible because of special functional requirements, for example, gloves in gloveboxes.

A-2. NRC Regulatory Guide 3.20

PROCESS OFF-GAS SYSTEMS FOR FUEL REPROCESSING PLANTS

A. INTRODUCTION

Section 50.34 of 10 CFR Part 50, “Licensing of Production and Utilization Facilities” requires, among other things, that each applicant for a construction permit for a production or utilization facility, including fuel reprocessing plants, include the principal design criteria for the facility. Paragraph (f) “Environmental Report,” of §50.30. “Filing of Applications for Licenses; Oath or Affirmation,” requires that an application for a construction permit or operating license for a fuel reprocessing plant be accompanied by an Environmental Report. Appendix D, “Interim Statement of General Policy and Procedure: Implementation of the National Environmental Policy Act of 1969 (Public Law91-190)”[m] to 10 CFR Part 50 requires, among other things, that the Environmental Report include a discussion of the status of compliance of the facility with applicable environmental quality standards and requirements. This regulatory guide provides information relative to establishing principal design criteria for equipment associated with process off-gas treatment systems for fuel reprocessing plants.

B. DISCUSSION

Process off-gas systems, i.e., vessel and dissolver off-gas systems are designed to confine hazardous chemical or radioactive materials evolved during process operations and radioactive waste storage and to reduce their concentration of gaseous process effluents to levels as low as practicable.[n] Usual constituents of process off-gas systems are gas collection systems, condensate removal systems, sampling and chemical monitoring systems, and control and instrumentation systems.

Process off-gas systems are generally located within building structures designed to prevent uncontrolled release of radioactive materials under postulated accident conditions. Although those portions of off-gas systems so located do not require designs which duplicate this capability to withstand postulated accidents, off-gas systems should be designed to work with building structures (confinement systems) and building ventilation systems to prevent uncontrolled releases of radioactive material. Those portions of off-gas systems not enclosed in protective structures should themselves be designed against failures which could cause uncontrolled release of radioactive materials under all normal operating loads plus those loads attributable to natural phenomena such as earthquakes and tornadoes postulated in the design criteria. Accident conditions postulated should also include equipment and utility failures and failures due to engineering or operating errors.

To meet reliability needs during postulated accident conditions, off-gas systems are generally equipped with duplicate equipment items designed to maintain critical functions, e.g., installed spare exhaust blowers and equipment that will automatically switch to emergency utility systems upon loss of prime utility source to assure off-gas flow.

Collection lines and off-gas treating equipment are exposed to cell atmospheres and decontamination solutions as well as the collected fluids. To assure system reliability, the materials used in lines and equipment must be nonflammable, resistant to heat and the corrosive effects of the collected gases and the strong chemicals used for equipment decontamination. Stainless steel is usually employed for process

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[m] On November 1. 1973, the Atomic Energy Commission published in the Federal Register (38 FR 30203) proposed amendments to its regulations which would place the provisions of Appendix D. to 10 CFR Part 50 into a now Part 51 to be entitled "Licensing and Regulatory Policy and Procedures for Environmental Protection."

[n] Guidance on "As Low As practicable" as stated in 10 CM Part 20 is being developed. This may include examination of krypton and tritium levels in process effluents.

lines and vessels, rather than plastics, which may be subject to radiation, fire, or thermal damage, or carbon steel, which may be subject to corrosive attack by process materials or decontamination solutions.

Off-gas systems function to handle both condensable and non-condensable gases generated in process operations and waste storage. The condensable component of the off-gas consists primarily of steam and water vapor. To minimize the load on the off-gas system and to increase the effectiveness of off-gas cleanup operations, condensables are generally removed from the off-gas stream by condensers and stack drains and then recycled to the process as water. Amounts of condensable in excess of process needs may be discharged as liquid waste to surface waters or revaporized and discharged to the atmosphere. The choice of discharge mode for excess condensables is generally determined by site factors such as meteorology, hydrology, and demography. Regardless of the mode of discharge, sampling and monitoring of all effluents is necessary to assure effective process operation and compliance with regulations concerning release of these effluents.

Off-gases are further treated by one or more mass transfer devices, as necessary, to meet release limits for both radioactive and noxious materials. Devices used often include caustic scrubbers, acid scrubbers, activated carbon adsorbers, and zeolite adsorbers to remove iodine, and water scrubbers to remove oxides of nitrogen. Performance data for these devices have been published.[o]

Particulate removal equipment with high collection efficiency is needed to satisfactorily clean gases prior to discharge to the environs. Various types of filters are usually used in this service. To assure reliable process operation, such filters should be resistant to fire or be preceded in the process train by fire-suppressing equipment. Filters and adsorbents such as zeolites can be damaged by liquids in the off-gas. To prevent such damage, filters and adsorbers may be preceded by heaters or by electrical or steam traced lines which maintain the off-gas above its dewpoint. ORNL-NSIC-65, “Design, Construction and Testing of High Efficiency Air Filtration Systems for Nuclear Application,”[p] offers a comprehensive review of air filtration systems. This document is not a standard but a guide which suggests several design alternatives.

Coordination of designs for the off-gas systems with the designs for the confinement systems and building ventilation systems is important to assure complementary functioning of these systems. These systems work together to keep process vessel pressures lower than pressures in surrounding process areas thereby limiting leakage of contaminated gases into process cells and thence into potentially inhabited plant areas.

C. REGULATORY POSITION

1. All parts of the process off-gas system should be designed:
    a. To limit the release of radioactive materials during normal operation to the levels stated in 10 CFR Part 20.
    b. To limit the release of noxious materials to comply with Federal and State statutes and implementing regulations imposed by Federal, State, and regional agencies.
    c. To withstand postulated accident conditions to the extent that uncontrolled release of radioactive material to the environs is prevented or to be enclosed within a structure designed to withstand postulated accident conditions to the same extent. Accident conditions postulated should include earthquake, tornado, equipment and utility failure, and engineering or operating error. The pertinent quality assurance requirements of Appendix B to 10 CFR Part 50 should be applied to all activities affecting the safety-related functions of these structures, systems, and components.

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[o] Aqueous Processing of LMFBR Fuels-Technical Assessment and Experimental Program Definition." pp. 167-186. USAEC report ORNL4436, Oak Ridge National Laboratory, June 1970.

[p] C. A. Burchsted and A. D. Fuller, "Design. Construction and Testing of High-Efficiency Air Filtration Systems for Nuclear Application." ORNL-NSIC-65, Oak Ridge National Laboratory, January 1970.

d. With adequate duplicate process and support equipment to maintain system safety functions in the event of any single failure or during maintenance operations.

e. To resist fire, thermal, effects, and the corrosive effects of cell atmospheres, decontamination solutions, and collected gases to the extent necessary to maintain safety-related functions.

f. To permit inspection, maintenance. and testing of systems and components that have safety-related functions to assure their continued functioning for the life of the facility.

2. The gas collection equipment should be designed to:

a. Collect gases near points of generation and conduct them in closed piping systems to treatment systems for the removal of hazardous chemical or radioactive materials.

b. Operate at negative pressures relative to surrounding cells where practical.

c. Prevent header flooding and unsafe accumulation of fissionable materials by sloping collection piping to drain to appropriate process vessels, by use of condensers and knockout pots, and by use of vessel overflow lines.

d. Minimize entrainment into collection headers, thereby preventing unsafe accumulation of fissionable materials, by sizing vessel off-gas lines to provide low gas velocities, by providing deentrainment devices, and by separating vessel off-gas lines from other vessel lines such as those receiving jet or pump discharge streams.

e. Limit spread of contamination by preventing backflows from radioactive to less radioactive areas by use of liquid seals on cold chemical addition lines, by providing top entry of off-gas branch lines into headers, and by providing pressure-relief devices to guard against pressure increases due to flow blockages or gas flows in excess of design specifications.

f. Minimize radiation exposure to plant personnel by locating process piping containing radioactive material away from areas frequently occupied by plant personnel or by providing local biological shielding.

3. The chemical treatment equipment should be designed to:

a. Remove both radioactive and noxious gaseous contaminants predictably and effectively from process off-gases. These contaminants include iodine, organic and inorganic iodine species, and other hazardous materials.

b. Provide protection to zeolite adsorbers against moisture damage.

4. The particulate removal equipment should be designed to:

a. Remove radioactive particulate contaminants predictably and effectively from process off-gas.

b. Resist fire and be located downstream from fire-suppressing process devices or be equipped with fire detection and extinguishing devices. Fire-suppressing devices may consist of other process units, such as wet scrubbers used for chemical treatment, or may be systems designed specifically for fire protection.

c. Resist or be protected from condensate damage by maintaining system temperatures above the dewpoint of the gas and by providing low-point traps and drains on supply headers.

5. The exhaust equipment should be designed:

a. To provide motive power adequate to overcome off-gas system head and maintain prescribed off-gas system pressures to prevent backflow of contaminated gases into potentially inhabited areas.

b. With emergency utilities provided automatically on failure of prime utility source.

6. The sampling and chemical monitoring equipment should provide:

a. Sampling points for off-gas on each chemical removal device and at the process off-gas system inlet and discharge points.

   b. Ports for testing filter efficiency on each safety-related stage of filtration.
   c. Redundant continuous monitoring devices adequate to measure and record overall effluent radioactivity and system performance at point of process off-gas system discharge to the environs. These devices may be stack monitors which monitor combined plant discharges.
   d. Alarms in a continuously occupied control room to indicate safety-related abnormal conditions.

7. Control and instrumentation systems should be designed to:
   a. Control automatically or facilitate manual control of all safety-related process parameters.
   b. Indicate or record all safety-related process parameters in a regularly occupied control room. Parameters include such items as fluid levels, pressures, temperatures, and radiation levels.
   c. Alarm all abnormal safety-related parameters in a continuously occupied control room.
   d. Switch in safety-related standby process devices automatically when needed.
   e. Switch safety-related process devices to standby power supplies automatically when needed.

A-3. NRC Regulatory Guide 3.32

GENERAL DESIGN GUIDE FOR VENTILATION SYSTEMS FOR FUEL REPROCESSING PLANTS

A. INTRODUCTION

Section 50.34, “Contents of Applications; Technical Information,” of 10 CFR Part 50, “Licensing of Production and Utilization Facilities,” requires, among other things, that each application for a construction permit for a production or utilization facility, including fuel reprocessing plants, include the principal design criteria for the facility. At fuel reprocessing plants, a principal risk to health and safety is the uncontrolled release and dispersal of airborne radioactive material. Structures, systems, and components important to safety in a fuel reprocessing plant include, among other things, features designed to prevent, limit, or mitigate the release of radioactive material. These features include protection by multiple confinement barrier and systems, ventilation systems, and off-gas systems.

Regulatory Guide 3.18, “Confinement Barriers and Systems for Fuel Reprocessing Plants,” provides information relative to establishing principal design criteria for confinement barriers and systems that will minimize the amount of radioactive material released to the environment or to areas normally occupied by personnel. Regulatory Guide 3.20, “Process 0ffgas System for Fuel Reprocessing Plants,” provides information relative to establishing principal design criteria for equipment associated with process off-gas treatment systems for reprocessing plants. The ventilation systems for a fuel reprocessing plant are designed to confine, channel, and control airborne radioactive contaminants and provide normal ventilation functions. Many acceptable bases for the design of ventilation systems for fuel reprocessing plants are available and have been used. These bases can differ slightly because there has not been sufficient guidance toward standardization. In the interest of standardization, this guide describes bases acceptable to the NRC staff for the design of ventilation systems for fuel reprocessing plants.

B. DISCUSSION

Ventilation systems for fuel reprocessing plants are designed to supply properly conditioned air to occupied and unoccupied areas, ensure that air is confined to prescribed flow paths for discharge via a final filter or treatment system and stack, and that proper monitoring and filtration or treatment are provided. Ventilation systems for current generation fuel reprocessing plant usually consist of through air supply and exhaust air systems together with associated air filters, fans, dampeners, ducts, monitoring equipment, and control instrumentation. Air filters can include medium efficiency prefilters and high-efficiency filters such as sand filters, fiberglass packs with deep bag polishing units, and HEPA (high-efficiency particulate air) filters.

Fresh air is drawn into the plant and conditioned by the air supply system and distributed to various zones occupied by personnel. Part of the air is directed from these zones into areas of greater potential for contamination but also accessible to personnel, and thence into process zones. Air from process area zones is removed through a ventilation exhaust and filtration system which discharges through a stack to the environment. Provisions may be made for alternative release points downstream of final exhaust air filters in the event that flow through a stack is blocked due to structural failure.

Part of the ventilation air for normally occupied zones may be recycled to the air supply system through recirculating air systems. Independent recirculating air systems may be used in selected areas of the plant having particular occupancy requirements such as control rooms or control areas that are designed to permit occupancy to operate the plant safely under normal conditions and to maintain the plant in a safe condition under accident or other abnormal conditions.

Ventilation systems will be subject to variations in operating temperatures and pressures and to environmental conditions associated with normal operation, maintenance, plant shutdown, and testing. They may also be subject to effects of natural phenomena such as seismic motion and floods and of missiles, fire, explosion, and other accidents.

The systems must continue to perform their safety functions effectively under all conditions by confining radioactive or other potentially dangerous materials. They should be designed to ensure that the concentration of radioactive materials in the effluent gases is as far below the limits specified in 10 CFR Part 20 as practicable.

The continuity of necessary ventilation can be ensured by means such as standby equipment and fail-safe control systems. The ability of the systems to perform their safety functions effectively can be ensured by periodic testing of safety-related components during normal operation of the systems to demonstrate their ability to perform at design efficiency and to verify availability for emergencies.

C. REGULATORY POSITION

Ventilation systems for a fuel reprocessing plant should ensure the confinement of hazardous materials during normal or abnormal conditions, including natural phenomena, fire, and explosion. The release of radioactive material to the environment or to an area in which levels of radioactivity are normally sufficiently low to permit personnel access should be reduced to a level as low as practicable in accordance with 10 CFR Part 20

1. General Safety

   a. Ventilation systems should be designed to confine radioactive materials as close to the point of origin as practicable and prevent uncontrolled release of radioactive aerosols, noxious fumes, and vapors into rooms and areas normally occupied by personnel.

   b. Confinement of radioactive materials should be provided by multiple-zone confinement barriers and systems. Negative pressure differentials should be maintained between building confinement zones and also between the confinement zones and the outside atmosphere to ensure that air flow is from zones of lesser potential for contamination to zones of greater potential for contamination. Features of confinement barriers and systems, including pressure monitoring and alarm requirements, are discussed in Regulatory Guide 3.18.

   c. Ventilation systems should be designed so that the failure of any one component (equipment or control device) will not affect the continuous operation of the ventilation systems. Ventilation systems and components should have fail-safe features with provision for alarm indication.

   d. Onsite emergency power should be provided to operate the ventilation systems and components, including instruments and controls, important to safety. Ventilation systems should be capable of operating during normal power outage at capacities required to maintain confinement of contaminants. The onsite emergency power sources and the electrical distribution circuits should have independence and testability to ensure performance of their safety functions assuming a single failure.

   e. The ventilation systems should be designed to withstand any credible fire and explosion and continue to act as confinement barriers. The ventilation systems should be constructed of fire-resistant materials of construction and should include fire-resistant filters, heat and smoke detectors, alarms, heat removal devices, fire-suppression equipment, and fire doors and dampers or other proven devices to restrict the spread of fires. The design of the fire protection system should include provisions to protect against adverse effects in the event of operation or failure of this system. For example, a drain system of critically safe geometry should be provided to prevent a criticality Incident in the event of water discharge on activation of a water spray heat removal system or in the event of water leakage on failure of a heat removal system component such as a spray nozzle while the system is not in operation.

f. Ventilation systems should be capable of operating during a fire in the areas they ventilate and safely handle products of combustion through appropriate ventilation channels. A supply air system should remain operational; however, the option to discontinue air supply to the involved space or spaces should be maintained. Any system that may be shut down should be protected against backflow.

g. The materials of construction for the ventilation systems should be fire-resistant to protect against fires occurring inside or outside the systems. All filters should be of a fire-resistant type and, where applicable, listed by Underwriters' Laboratories (Ref. 1) or the Factory Mutual Research Corporation (Ref. 2). Filters and exhaust fans, especially a final filter plenum and exhaust fan enclosure, should be so located as not to be exposed to the direct effects of fire or explosion in the operating areas. Smoke and heat detectors listed by the Underwriters' Laboratories (Ref. 3) or the Factory Mutual Research Corporation (Ref. 2) should be provided in the ventilation systems.

h. Fire- and smoke-suppression equipment should be so located as to ensure that the integrity of final high-efficiency filters or filter systems is not degraded. Spark and flame arresters and isolation valves may be used at filter installations in intermediate stages of effluent cleaning. Where appropriate, a heat removal system should precede a high-efficiency filtration system serving as a final means of effluent cleaning (see regulatory position C.4.a(6)).

i. Normally, plant design and operating procedures should limit quantities of combustibles. Where sources of combustible solvents, gases, and vapors are identified or postulated to be present in a ventilation system under normal or abnormal conditions, suitable continuous monitoring systems should be employed. These monitoring systems should sound audible alarms and display visual alarm indications to operating personnel when the prescribed safe limits for combustible gas and vapor mixtures are reached. The monitoring and alarm system itself should not introduce an ignition source and should not affect the confinement integrity of the ventilation system. The monitoring and alarm system should be designed for in-place calibration and testing.

j. The ventilation systems should be designed to withstand tornado conditions without loss of confinement capability resulting from mechanical damage to the system or components or from the reduced ambient pressure at the intake and exhaust openings of the building. Protection against missiles should be provided for the intake and exhaust openings, as well as for any exposed (outdoor) ductwork or equipment between the intakes and exhaust filters.

k. Components of the ventilation systems should be designed to withstand the effects of earthquakes and remain functional to the extent that they will prevent the uncontrolled release of radioactive materials to the environment.

2. Occupied Area Ventilation Systems

a. Supply air should be properly conditioned and distributed, where practical, in a downward flow pattern to the potentially contaminated areas of the facility.

b. Outside makeup air supply units should be protected from the weather and should be so located as to minimize potential for intake of stack discharge gases. Inlets should be so arranged as to minimize the effects of high winds, rain, snow, ice, and debris on the operation of the system. Heaters may be necessary in areas where icing can cause significant supply filter damage. Trash screens or other proven devices should be provided over supply air inlets to protect air treatment equipment and filters from debris.

c. Where air from occupied areas is directed to contaminated or potentially contaminated areas, consideration should be given to passing this air through suitable filters to prevent backflow of particulate contaminants.

d. Part of the air in normally occupied areas may be recirculated to reduce thermal loads. Air containing noxious, toxic, or corrosive gases and vapors should not be recirculated. Recirculating air systems should be equipped with adequate air cleaning equipment to ensure the maintenance of air quality in occupied areas. Provisions should be made for monitoring the recirculated air. Upon any indication that the limits for radioactivity in occupied areas specified in 10 CFR Part 20 or in the technical specifications have been reached, the air in the recirculating system should be diverted to the once-through exhaust ventilation system for discharge through a final filtration system.

e. Recirculating air systems independent of ventilation systems may be used in selected areas of the plant having particular occupancy requirements such as control rooms or control areas.

f. Air locks should be provided where frequent entry between personnel occupancy and limited access areas is necessary and where air flow must be maintained in one direction.

3. Process Area Ventilation Systems

a. Fire-resistant medium-efficiency prefilters should be used in process area ventilation systems to remove the bulk of particulate matter in the air streams and thus limit the loading of the high-efficiency filters installed in the exhaust air systems. Each prefilter should have an atmosphere dust spot efficiency rating (Ref. 4) of approximately 80% or better.

b. Where necessary, appropriate means and procedures should be provided in addition to the process off-gas treatment systems (see Regulatory Guide 3.20) to protect final HEPA filter systems from exposure to wetting or deleterious chemical attack.

c. The ventilation systems should maintain minimum air velocities of 120 linear feet per minute (design velocity should be ~150 linear feet per minute) through all process area openings such as fume hood doors and process cell covers to prevent significant reverse flow of contaminated air.

d. Air or inert gas should enter each ventilated glove box through at least one fire-resistant HEPA filter and be discharged through at least one fire-resistant HEPA filter to exhaust ductwork leading to a final filter system (see regulatory position C.4.a(6)). The inlet filter prevents any backflow of contaminants into the work areas, and the outlet filter minimizes contamination of the exhaust ductwork.

e. Consideration should be given to recirculation of exhaust air or inert gas in glove boxes to minimize release of radioactive particulates to the biosphere. If recirculation is used, the exhaust gas from these enclosures should be filtered through at least one fire-resistant HEPA filter before being recirculated.

f. The features described in regulatory position C.7 of Regulatory Guide 3.12, “General Design Guide for Ventilation Systems of Plutonium Processing and. Fuel Fabrication Plants,” addressed specifically to glove boxes for plutonium processing and fuel fabrication plants are also generally applicable to glove boxes for fuel reprocessing plants.

4. Exhaust Ventilation and Filtration Systems

a. HEPA Filter Systems

(1) Each exhaust filter housing should have a rigid mounting frame for the filter. The leakage of the complete housing structure should be as low as possible from outside to inside and zero from inside to outside or across the filter-sealing barrier (exclusive of the filter). The filter-sealing barrier should be made leak-tight without resorting to sealing tapes or caulking.

(2) Where filter access openings in these housings are provided for filter removal, the configurations should permit filter removal and replacement with minimum exposure to personnel performing this task and with minimum release of contaminants outside of the housing.

(3) The filter housings should be equipped with necessary test ports to permit reliable in-place testing of all filter stages with dioctyl phthalate (DOP).

(4) The minimum instrumentation for each filter housing should include a device or multiple devices to indicate filter resistance and airflow rate.

(5) Where filter systems are designed for replacement, isolation valves should be so located that a bank of filters can be completely isolated from the ventilation systems during filter replacement operations.

(6) A heat removal system and a spark arrester should precede each HEPA filtration system serving as a final means of effluent cleaning. If a cooling spray such as a water spray system is used for heat removal, it should be followed by a combination spark arrester/ moisture separator screen to also remove entrained droplets, thereby protecting filters from plugging and damage. A roughing filter and a wire mesh protective screen should be mounted behind these components to remove the bulk of any draft-carried debris and thus avoid loading the HEPA filters installed downstream.

The cooling spray system should operate automatically (with a manual override) when an abnormal heat rise in the cooling chamber inlet is indicated by detectors in exhaust ducts feeding the filter plenum. A manually operated valve actuating the spray system should also be provided as a backup. If a drain system is installed to prevent accumulation of liquid in the plenum, the collection tanks should be of critically safe geometry.

(7) The filtered air should be discharged to the environs through a stack of sufficient height to reduce close-in ground-level concentrations of radioactive or other potentially dangerous contaminants. Provisions should be made for an alternative release point in the event that flow through the stack is blocked because of structural failure.

(8) The stack should be located sufficiently distant from other facilities that structural failure would not result in damage to any process systems or structures important to safety.

(9) HEPA filters used in intermediate and final filtration systems should be designed to military specifications MIL-F-51068D (Ref. 5) and MIL-F-51079B (Ref. 6) and should satisfy the requirements of UL-586 (Ref. 7).

(10) HEPA filter systems should be tested after filter installation using a “cold DOP” test. Acceptance should be based on an efficiency of 99.95% or better for DOP having a light-scattering mean diameter of approximately 0.7 micron. Regular in-place testing of both on-line and standby filter installations should be performed because system deterioration can take place even when the installations are not being used. Test procedures used should comply with the recommendations contained in ORNL-NSIC-65, “Design, Construction and Testing of High-Efficiency Air Filtration Systems for Nuclear Application,” (Ref. 8) and in Regulatory Guide 3.2, “Efficiency Testing of Air-Cleaning Systems Containing Devices for Removal of Particles.”

b. Other Filter Systems

(1) Final filtration systems incorporating high-efficiency filters other than, HEPA filters, such as packed glass wool or packed sand units, should have equivalent efficiency and resistance to fire.

(2) Packed glass wool and packed sand filter systems should be tested prior to operation using a “hot DOP” test and provisions should be incorporated in the installations for periodic in-place “cold DOP” testing (see Regulatory Guide 3.2).

(3) HEPA filter systems used in combination with other filter systems should meet the recommendations of paragraph 4.a(10) above.

5. Fans

a. Installed spare fans and isolation dampers should be provided for the ventilation systems. When any one fan is inoperative in a system, a backflow damper should automatically isolate the idle fan from the system. Standby fans should automatically start and have sufficient capacity to maintain minimum system air flow.

b. Alarms should be provided in one or more continuously occupied control areas to indicate malfunction of each ventilation fan (see regulatory position C.6.f).

c. Supply air fans should be interlocked with an exhaust air plenum pressure sensor to prevent supply fan operation unless the exhaust fans are running. This will prevent pressurization of any potentially contaminated area should exhaust ventilation fail.

d. Emergency power should .be supplied automatically to fans in the event of failure of the normal power supply (see regulatory position C.1.d).

6. Ventilation System Construction and Layout

a. The materials of construction for the ventilation systems should be carefully selected according to such safety considerations as strength to withstand accident conditions; corrosion resistance, particularly when associated with chemical processes; fire resistance; long operating life to avoid frequent replacement of contaminated equipment; and smooth surface finish to aid in decontamination.

b. Ducts and housings should be designed, fabricated, and erected with a minimum of ledges, protrusions, and crevices that can collect dust and moisture or that could impede personnel or create a hazard in performance of their work. Duct runs and flow distributors should ensure uniform, representative air flow past monitoring and sampling stations as well as through filter installations.

c. The design and construction should provide for convenient inspection, maintenance, decontamination, and replacement of critical components such as filters, fans, and system controls.

d. Housings, filter mounting frames, and ducts should be designed to withstand system pressure changes without distortion, vibration, fatigue, or yielding of such magnitude that leakage or bypassing of the filters results.

e. Supply, recirculation, and exhaust ducting should incorporate manual and automatic dampers and controls to distribute and regulate the movement of air in each room, area, and ventilation system and to prevent possible backflow in case of pressure reversal.

f. The ventilation systems should be appropriately instrumented to read out and alarm in one or more continuously occupied control areas. These areas should be designed to permit occupancy and to permit actions to be taken to operate the ventilation systems safely during normal or abnormal conditions.

7. Ventilation System Testing and Monitoring

a. Provisions should be made so that components of ventilation systems can be tested periodically for operability and required functional performance. They should include capability for periodic measurement of air flows in exhaust ducts and in or at equipment, hoods, and glove boxes.

b. The capability should be provided to test, under conditions as close to design as practicable, the operating sequence that would bring ventilation systems into action, including the transfer to alternative power sources, and the capability for delivering design air flow.
Regulatory Guide 3.22, “Periodic Testing of Fuel Reprocessing Plant Protection System Actuation Functions,” provides information relative to periodic testing of protection system actuation functions.

c. Exhaust ducts and stacks that may contain radioactive contaminants should be provided with a fixed sampler and a continuous monitor [Continuous Air Monitoring System (CAMS)] that provides a record of plant effluents. The probes for sampling purposes should be designed for isokinetic sampling and located to obtain representative samples. Each monitoring system should be connected to an emergency power supply (see regulatory position C.1.d). The continuous stack sampler should alert cognizant personnel in continuously occupied control rooms or areas through an audible and visual annunciator if the airborne radioactive effluents reach prescribed limits. Features of sampling and monitoring equipment for process off-gases are discussed in Regulatory Guide 3.20.

d. Air monitoring and warning systems (including CAMS) should be installed in areas where radioactive material is handled. Air-sampling heads should provide a representative sample of the potential airborne radioactivity being breathed. Consideration should be given to locating continuously operating exhaust samplers after each stage of high-efficiency filtration or, as a minimum, providing special connections to allow probes to be inserted for sampling.

8. Quality Assurance Program

a. A quality assurance program should be established for the design, construction, testing, operation, and maintenance of all structures, systems, and components addressed in this guide in accordance with the criteria in Appendix B to 10 CFR Part 50. Regulatory Guides 3.3, “Quality Assurance Program Requirements for Fuel Reprocessing Plants and for Plutonium Processing and Fuel Fabrication Plants,” and 3.21, “Quality Assurance Requirements for Protective Coating Applied to Fuel Reprocessing and to Plutonium Processing and Fuel Fabrication Plants,” describe, respectively, acceptable methods of complying with Appendix B with regard to overall program requirements and to requirements for protective coatings.

D. IMPLEMENTATION

The purpose of- this section is to provide information to applicants and licensees regarding the NRC staff's plans for utilizing this regulatory guide.

Except in those cases in which the applicant proposes to use an acceptable alternative method for complying with specific portions of the Commission’s regulations. the method described herein will be used in the evaluation of submittals for construction permit or operating license applications docketed after May 31, 1976.

If an applicant wishes to use this regulatory guide in developing submittals for an application docketed prior to May 31, 1976, the pertinent portions of the application will be evaluated on the basis of this guide.

REFERENCES

1. Underwriters' Laboratories Building Materials List (latest edition). Copies may be obtained from Underwriters' Laboratories, Inc., 207 East Ohio Street, Chicago, Illinois 60611.
2. Factory Mutual Approval Guide (latest edition). Copies may be obtained from Factory Mutual Research Corporation, 1151 Boston-Providence Turnpike, Norwood, Massachusetts 02062.
3. Underwriters' Laboratories Fire Protection Equipment List (latest edition). Copies may be obtained from Underwriters' Laboratories, Inc., 207 East Ohio Street, Chicago, Illinois 60611.
4. ASHRAE (American Society of Heating, Refrigerating and Air-Conditioning Engineers) Standard 52-68, "Method of Testing Air Cleaning Devices Used in General Ventilation for Removing Particulate Matter," Section 9. (Copies may be obtained from American Society of Heating, Refrigerating and Air-Conditioning Engineers, Inc., United Engineering Center, 345 East 47th Street, New York, New York 10017.)
5. MIL-F-51068D, "Filter, Particulate, High-Efficiency, Fire-Resistant," Military Specification. Copies may be obtained from Commanding Officer, Naval Publications and Forms Center, 5801 Tabor Avenue, Philadelphia, Pennsylvania 19120.
6. MIL-F-51079B, "Filter Medium, Fire-Resistant, High-Efficiency," Military Specification. Copies may be obtained from Commanding Officer, Naval Publications and Forms Center,'5801 Tabor Avenue, Philadelphia, Pennsylvania 19120.
7. Underwriters' Laboratories Standard UL-586, "High-Efficiency Air Filtration Units," (also designated ANSI B132.1-1971). Copies may be obtained from Underwriters' Laboratories, Inc., 207 East Ohio Street, Chicago, Illinois 60611.
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Appendix B

B-1. Summary of Contemporary Commercial Reprocessing Facilities

Nuclear Fuel Services Company, West Valley, New York, was operated by W.R. Grace Company from 1966 to 1971. It was the only commercial PUREX plant to be operated in the U.S. The plant reprocessed fuels from commercial LWRs and the Hanford N-Reactor. The nominal design capacity was 300 MTHM per year. The plant was shut down in December 1971 for reconstruction to nearly triple its capacity. Work on this effort continued until 1976 when the company suspended operations [57].

Midwest Fuel Recovery Plant, Morris, Illinois, was built by General Electric between 1970 and 1974 but never operated with irradiated fuels. Early testing revealed a flowsheet design flaw, and the plant was declared inoperable in 1975. Reprocessing was based on the Aquafluor process, which is a combination of solvent extraction coupled with uranium fluoride volatility. The nominal design capacity was 300 MTHM per year [58].

Barnwell Nuclear Fuel Plant, Barnwell, South Carolina, was built by Allied General Nuclear Services between 1970 and 1975 but never operated with irradiated fuels. Reprocessing was based on PUREX with a nominal design capacity of 1500 MTHM per year [59, 60].

Exxon Nuclear Fuel Recovery and Recycling Center, Oak Ridge Reservation, Ok Ridge, Tennessee. Construction permit application submitted to the NRC by the Exxon Nuclear Company, Inc., in 1976. The nominal design capacity was 2100 MTHM per year. The project never came to fruition [61]. Getty Oil Company, Atlantic Richfield Company, and Allied-Gulf Corporation were developing similar plans to construct commercial reprocessing facilities at that time.

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