Requirements and Conceptual Design of Off-gas Systems for the Reprocessing of Metallic Fuels

Fuel Cycle Research & Development

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SUMMARY

An assessment has been conducted to determine how key regulations regarding volatile radionuclide emissions to the atmosphere may apply to the off-gas streams associated with electrochemical reprocessing. The scope of this assessment was based upon a generic electrochemical reprocessing scheme with a throughput rate of 200 MTIHM/y applied to metallic fuel discharged from a sodium fast reactor (SFR), but the findings are able to be translated to other advanced nuclear scenarios as merited.

Air dispersion modeling was performed using the EPA CAP-88 model and evaluated the uncontrolled decontamination factors (DFs) that would be required to achieve regulatory compliance with the dose-based limits set forth by EPA regulation 40 CFR 190.10(a). These DFs were compared to those required by fuel cycle–based limits set forth by EPA regulation 40 CFR 190.10(b). Two theoretical sites with disparate climatological conditions were selected for air dispersion modeling (Idaho and Tennessee).

The radionuclides modeled included ^{3}H, ^{85}Kr, ^{129}I, and selected alpha-emitting transuranic isotopes (referred to here as ^{239}Pu-TRU$_{<1y}$). It was found that the fuel cycle-based limits in 40 CFR 190.10(b) are most restrictive for ^{85}Kr and ^{239}Pu-TRU$_{<1y}$, with DFs of 3 and 6.1E+09, respectively. The dose-based limit as derived from 40 CFR 190.10(a) could require mitigation of tritium in some scenarios, with an estimated DF of about 3 for the reference scenarios.

The fuel cycle-based limit for ^{129}I resulted in a DF of about 240 for the reference scenario. The need for iodine mitigation based on dose to the public depended upon the physical form of iodine as either particulate or vapor-phase species. Emission of iodine from the facility as a vapor necessitated DFs of about 2 but emission as a particulate would require DFs >6,000 to meet thyroid dose-based limits. Effects of physical form on needed iodine mitigation are significant, but the understanding of speciation of iodine both during electrochemical reprocessing and after release to the atmosphere is limited.

The electrochemical processing unit operations were evaluated to identify potential release points for the volatile radionuclides and to assess the potential for retention of the radionuclides within the process (thus decreasing the need for mitigation). Mitigation strategies for ^{3}H, ^{85}Kr, ^{129}I, and ^{239}Pu-TRU$_{<1y}$ were identified. In all cases, there are reasonably achievable pathways to regulatory compliance, although in some cases additional R&D is merited to verify the chemical speciation of these isotopes and to develop and demonstrate potential treatment technologies for this application.

Whether or not additional off-gas controls (beyond common operations such as HEPA filtration and oxygen and moisture control) are needed for any of these regulated or volatile radionuclides depends on the (a) type of facility (NRC-regulated or DOE), (b) used fuel process rate, (c) used fuel burnup and composition, (d) speciation and retention of volatile radionuclides in the process and in the cell gas cleanup system, (e) site-specific parameters such as location, meteorology, stack height, and site boundaries, and (f) levels of conservatism and safety factors used in assessing compliance to air emissions regulations.

Performance of this assessment revealed several areas where information is lacking or additional research is required in order to better determine if or what kinds of off-gas control might be needed. First, and most significantly, the understanding of the chemical speciation and physical form and partitioning of iodine during electrochemical processing operations is lacking and prevents the ability to accurately assess the potential iodine mitigation requirements. Future research in this area should be multifaceted and include thermodynamic modeling of iodine speciation in different process steps, experiments to quantify the kinetics of vapor-phase and melt-phase transitions, bench-scale experiments to determine the potential chemical and physical form of iodine emissions from the electrorefining process, and verification of iodine behavior with experiments utilizing operational facilities. Similarly, an improved understanding of iodine behavior in the environment after release from the facility stack will be required to refine dose estimations, as particulate and vapor-phase emissions result in significantly different doses to the MEI.

In the mitigation of specific radionuclides, several key topics merit additional R&D. First, the verification of tritium speciation and behavior during reprocessing and within a conceptual cell gas treatment system should be performed. Second, the development of alternative iodine sorbents should be pursued. Third, solid sorbents previously developed for noble gas separations should be tested in conditions that might be expected in this application.

The assessment conducted here provides a useful reference case for determining if and how off-gas emissions controls might be needed for electrochemical reprocessing of advanced reactor metallic fuels. The methodology used here can also be applied to emissions arising from other advanced nuclear technologies, including the operation of molten salt reactors and the reprocessing of advanced nuclear fuels. Importantly, the knowledge gaps identified as relating to the specific scenario evaluated (electrochemical reprocessing of metallic SFR fuel) will also be applicable to other advanced nuclear scenarios too, allowing for R&D to support multiple technologies. Future research should focus on fundamental chemistry of volatile radionuclide release from electrochemical processing and subsequent adsorption of volatile radionuclides so that as additional nuclear technologies approach deployment the scientific barriers to implementation will be reduced.

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FIGURES

TABLES

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ACRONYMS

AgZ	silver mordenite
AgZ-PAN	silver mordenite in a polyacrylonitrile-based engineered form
CAP-88	EPA model Clean Air Act Assessment Package – 1988
CWF	ceramic waste form
DF	decontamination factor
DOE	US Department of Energy
EPA	Environmental Protection Agency
ER	electrorefiner
FCF	Fuel Cycle Facility
HEPA	high-efficiency particle air
IFR	Integral Fast Reactor
INL	Idaho National Laboratory
MEI	maximum exposed individual
MOX	mixed oxide (UO_2–PuO_2)
MWF	metal waste form
ORNL	Oak Ridge National Laboratory
PWR	pressurized water reactor
R&D	research and development
UNF	used nuclear fuel
UOX	uranium oxide (UO_2)

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REQUIREMENTS AND CONCEPTUAL DESIGN OF OFF-GAS SYSTEMS FOR THE REPROCESSING OF METALLIC FUELS

1. INTRODUCTION

The reprocessing of irradiated nuclear fuel is a key element in the deployment of advanced nuclear technology. Recovery of key fissile isotopes from used nuclear fuel (UNF) is required to operate a closed nuclear fuel cycle, in which the amount of energy extracted from thorium, uranium, or plutonium-containing nuclear fuels is maximized. Beyond this, the reprocessing of UNF can achieve other objectives, such as recovery of valuable fission products for diverse uses (including the recovery of isotopes for medical use or for use in space applications). Minimization of the volume of high-level waste is an additional benefit in the reprocessing of UNF. Experts have identified scenarios that include operation of fast reactors with continuous recycle of nuclear fuel as alternatives to the currently implemented once-through nuclear fuel cycle followed in the US (Wigeland et al. 2014). Some of these alternatives use metallic fuel within a fast reactor.

Recovery of fissile material (uranium and plutonium) from uranium oxide–based nuclear fuels has historically proceeded through aqueous-based separations but recycle of some advanced nuclear reactor fuels may proceed more efficiently through other types of separations, including electrochemical separations. Electrochemical separations are especially well-suited for the reprocessing of metallic fuel.

Metallic fuel (or metallic alloy fuel, used synonymously in this document) may be used by a variety of nuclear reactor types. Historically, metallic fuel has been most closely associated with the deployment of large-scale sodium-cooled fast reactors (SFRs). However, as reactor design concepts have diversified (to include small modular reactors, micro-reactors and reactors for deployment in space), the applications for metallic fuel have expanded correspondingly. The GE-Hitachi PRISM design, an SFR using metallic fuel, is the basis of the cost estimates being developed in support of the US Versatile Test Reactor (Allen 2018). The primary constituent of metallic fuel is uranium, and the uranium may be alloyed with plutonium, zirconium, or a combination of both. The enrichment level of the uranium within the fuel will be dependent on the specific reactor design and may use high-assay low-enriched uranium. As compared with oxide-based fuels, metallic fuels have higher thermal conductivity and lower heat capacity. Cladding materials will be dependent on specific reactor design (taking into consideration the neutron spectrum, compatibility with the coolant material, and other factors) but stainless-steel cladding has been extensively examined for use in metallic-fueled SFRs.

The reprocessing of UNF by any means will liberate gaseous fission products from the irradiated fuel. Potential volatile elements include hydrogen, carbon, noble gases, and halogens, and these elements will distribute through chemical processing operations based on their individual chemical speciation and physical properties. Many of these volatile radionuclides will partition into the off-gas streams associated with fuel reprocessing. These off-gas streams arise from multiple sources, such as the sparging of liquids or molten salts, venting of process vessels, and sweeping or blanketing of operations conducted in an inert environment. Any venting of radionuclide-bearing process gas streams to the environment is governed by US regulations limiting the emitted radioactivity from nuclear facilities.

The applicable regulations include 40 CFR 190.10, 40 CFR 61, and 10 CFR 20 (EPA 2010a, EPA 2010b, NRC 2012). Extensive technical assessments have been performed to determine how these regulations may be applied to a reprocessing plant based on aqueous separations technology; these assessments were developed using detailed understanding of the chemical emissions of the processing technologies, the potential plant design, and the transport or dispersion of volatile radionuclides that may occur after emission from the facility (Soelberg 2008, Jubin et al. 2012a, Jubin et al. 2012b, Jubin et al. 2013, Jubin et al. 2014, Jubin et al. 2016). Similar assessments have not yet been performed for a facility using electrochemical reprocessing. Given the increasing interest in metallic fuels by advanced reactor developers it is important to extend these analyses to the electrochemical processing of metallic fuel.

A wide range of fast reactor types and concepts include SFRs, lead-cooled fast reactors, various gas-cooled reactors, and molten salt fast reactors. Different fuels and fuel types for these many different reactor types including metallic uranium, metallic uranium-transuranic (U-TRU), uranium oxide, nitride, carbide, mixed oxide, molten salt, and tri-structural isotropic (TRISO) fuels. Different claddings and fuel assembly types are used for these various fuel types, including no fuel assembly or cladding for molten salt fuels. This wide range of fast reactors, fuels, cladding, and fuel assemblies is beyond the practical scope of this report. This report focuses only on electrochemical reprocessing of advanced reactor metallic U-TRU fuel, as specified in the Fiscal Year 2020 Fuel Cycle Research and Development Milestones for Oak Ridge National Laboratory and Idaho National Laboratory. However, the analysis methodology used in this report, adapted from the methodology developed for analyzing off-gas control requirements for aqueous reprocessing, can also be used for reprocessing other used fuel types from other reactor types.

This report seeks to determine how current US regulations may limit the release of volatile radionuclides from an electrochemical-based UNF reprocessing facility. The analysis performed is based on a generalized hypothetical commercial electrochemical reprocessing concept operated within a facility that is regulated by the Nuclear Regulatory Commission (NRC). This allows for conclusions that may be broadly applied across multiple electrochemical reprocessing implementation scenarios. Other regulations can apply to other facilities, such as for Department of Energy (DOE) facilities. The potential for volatile radionuclide release from individual unit operations is assessed using empirical data where available. Air dispersion modeling is used to estimate the level of radionuclide mitigation (also termed control efficiency or decontamination factor [DF] in this report) that may be required. Since any future reprocessing facility may not match the assumed parameters of this hypothetical, generalized case, multiple scenarios are examined to assess the effects of throughput, volatile radionuclide speciation, meteorology and other aspects of the facility location, and facility design choices that can affect air dispersion and needed DFs. Potential mitigation strategies are identified and include recommendations for process design and off-gas capture, as well as identification of key knowledge gaps that may impact efficient volatile radionuclide management.

2. BACKGROUND

Three types of information provide a technical basis for the analysis described in this report. The first is the design and operational parameters that might be expected for an electrochemical reprocessing facility. Although the findings described in this report are not facility-specific, an examination of operating and conceptual facilities provides the basis for understanding the release of volatile radionuclides from the fuel into plant off-gas streams. The second type of information required is an understanding of how US regulations may be applied to the gaseous emissions of an electrochemical UNF reprocessing facility. For this, several detailed reports commissioned by the US DOE are summarized here. Finally, a description of the three key regulations that limit radionuclide emissions are included for reference.

2.1 Prior Experience with Electrochemical Reprocessing

Two pilot-scale electrochemical reprocessing facilities have been operated internationally, the first located at the Research Institute of Atomic Reactors in Dimitrovgrad, Russia, and the second associated within the Fuel Cycle Facility (FCF) at the Idaho National Laboratory within the US. The Research Institute of Atomic Reactors and other smaller-scale international facilities have been focused on the reprocessing of uranium and plutonium mixed oxide (MOX) fuel. The FCF was the processing facility associated with the Experimental Breeder Reactor-II (EBR-II) and was at the forefront of electrochemical reprocessing of metallic fuel. Other countries have made substantial investments in development of electrochemical reprocessing (including South Korea, France, and the United Kingdom) but these efforts were directed at the processing of oxide fuels. A full summary of international research and development (R&D) related to electrochemical reprocessing of UNF is available in NEA-OECD (2018). The review provided here focuses on domestic electrochemical reprocessing of metallic fuels, which is most applicable to the specified scope.

Multiple implementation concepts have been developed for the operation of metallic-fueled SFRs and associated reprocessing facilities. In some cases, a single reactor is envisioned as being sited with a dedicated fuel recycle facility (to include electrochemical recovery of fissionable material, waste stabilization, and fuel fabrication). Other scenarios have described multiple reactors sited with a single, relatively larger, fuel recycle facility. Some of the major implementation efforts, both conceptual and demonstration, are described here.

2.1.1 Fuel Cycle Facility

The FCF was designed to support the demonstration of a closed fuel cycle concept centered on the operation of an SFR (in this case, EBR-II) and associated fissile material recovery and subsequent fuel fabrication (Goff et al. 2011). After successful demonstration of this concept in the 1960s, the facility was redesigned and has housed significant R&D efforts focused on the development of electrochemical reprocessing. Some of these efforts supported the design of the Integral Fast Reactor (IFR) discussed in Section 2.1.2. Research performed at the FCF forms the basis of US experience with electrochemical reprocessing of metallic fuel. It currently houses the Mark-IV Electrorefiner, an electrorefiner (ER) designed to treat 16 kg fuel per batch and utilized in the disposition of irradiated sodium-bonded EBR-II driver fuel. Other unit operations within the FCF (now denoted the Fuel Conditioning Facility) include a cathode processor/salt distillation apparatus, metal casting furnace, and a fuel chopper.

2.1.2 Integral Fast Reactor and the GE PRISM Design

The IFR was conceived in some ways as the successor to EBR-II, heavily leveraging the achievements of EBR-II and its associated fuel recycle program in the conceptual approach of a co-located reactor and fuel reprocessing facility. The IFR's key design requirements included a sodium-cooled pool-type reactor, metallic fuel, and recovery of fissile components via electrochemical reprocessing for fuel refabrication. The IFR was intended to provide 1.4 GWe and would reprocess 19.3 MTIHM/y, corresponding to a fuel burnup of 99 GWd/MTIHM (Lineberry et al. 1985). IFR design requirements aligned with commercial initiatives of the time, including the GE PRISM reactor. The IFR project was discontinued before completion.

The PRISM reactor is still of commercial interest and remains under development by GE-Hitachi. GE-Hitachi deployment scenarios envision the electrochemical reprocessing of used PRISM fuel to recover fissile material. Such reprocessing would be performed co-located with the PRISM reactor as part of an "Advanced Recycling Center" (GE Hitachi Nuclear Energy 2020). The PRISM reactor is designed to generate 311 MWe.

2.1.3 Advanced Burner Reactor/Advanced Fuel Cycle Facility

The Global Nuclear Energy Partnership commenced in 2006 and within that framework the concept of an Advanced Fuel Cycle Facility to be sited in the US was developed. The Advanced Fuel Cycle Facility was envisioned as a multipurpose R&D facility and was explicitly intended to include electrochemical reprocessing demonstration capabilities on the scale of 1 MTIHM/y. It was designed to process a fraction of the fuel discharged from the conceptual Advanced Burner Reactor (with a reference design of plutonium-based metallic fuel), with the remainder processed using aqueous-based technology (Cahalan et al. 2007). The design effort was discontinued upon Global Nuclear Energy Partnership cancellation by the US DOE in 2009.

2.1.4 Summary and Assumptions regarding Facility Specifics

Demonstration efforts for the electrochemical processing of used metallic fuel generated by SFRs is limited, but several large-scale design efforts have been undertaken in the US and either abandoned before completion or not yet selected for commercial use. For this reason, the analysis undertaken in this report can only be based on a generic operational scenario. Conversations with current experts in the field indicate that an upper bounding throughput for a fuel reprocessing facility based on electrochemical reprocessing is likely in the range of 100–200 MTIHM/y, which could support five or more 1 GWe SFRs. The fuel to be processed is expected to be cooled 1–2 y. Required unit operations would likely include fuel disassembly and chopping, electrorefining, product purification, molten salt recycle, and waste form fabrication. The following assumptions are included in this report:

- The electrochemical processing facility throughput rate of 200 MTIHM/y was used in air dispersion modeling to estimate dose and dose-based DFs.
- The reference case fuel cycle assumed in this electrochemical reprocessing off-gas study is the continuous recycle of used metallic U/Zr fuel from an SFR to produce U/Zr and U/TRU product material for future fabrication of nuclear fuel. The fuel cycle is assumed to be at equilibrium.
- Initial fuel composition is assumed to be 70wt%U–20 wt% transuranic (TRU)–10 wt% Zr. The equilibrium composition of the TRU component of the used fuel for the system with a conversion ratio of 0.75 is 0.21% Np–17.7 wt% Pu–0.92 wt% Am–0.38 wt% Cm.
- The used fuel will be assumed to be cooled for 2 y before processing and will be assumed to have a burnup of 99.6 GWd/MT. Additional details on the spent fuel composition are provided in Section 3.

This system is representative of electrochemical reprocessing in other references including Lineberry 1985, DOE 2007, Law 2015, Vienna 2015, and Dixon 2019 and reflects one of the fuel cycles identified by Wigeland et al. (2014). This conceptual fuel cycle (referred to as EG-24 by Wigeland) is shown in Figure 1.

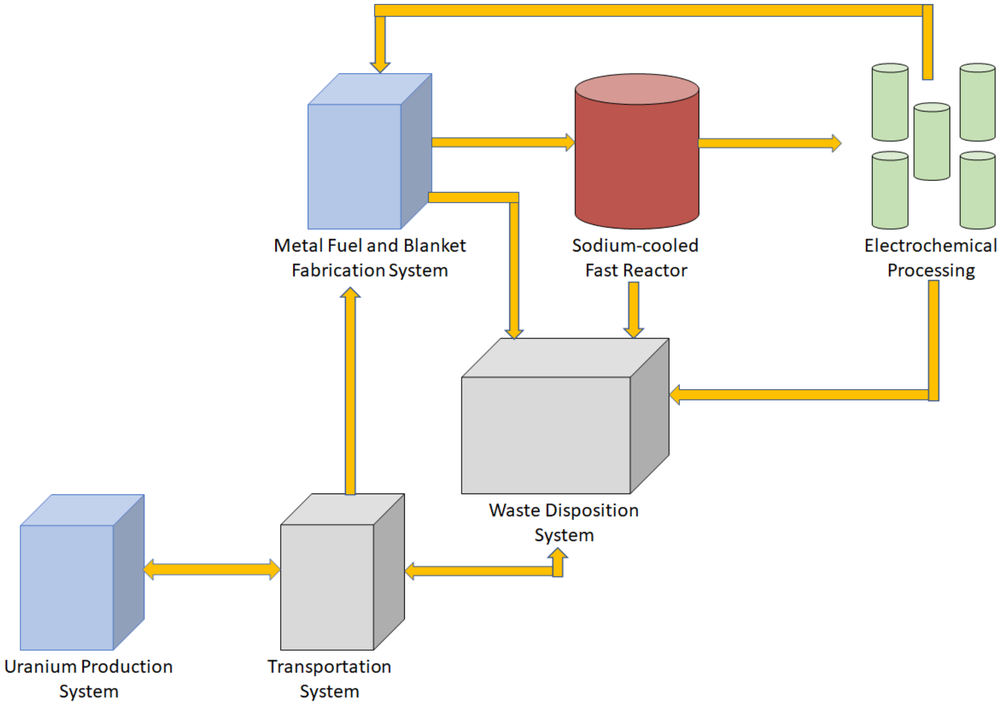

Figure 1: Electrochemical fuel separation system in a fast reactor recycle fuel cycle (adapted from Dixon et al. 2019).

2.2 Emissions Assessments Related to Nuclear Fuel Reprocessing

Several foundational reports have been issued assessing the extent of radionuclide mitigation required in the operation of an aqueous reprocessing facility in order to achieve compliance with US regulations (Jubin et al. 2012a, Jubin et al. 2014, Law et al. 2015). As this report builds on the techniques and findings of those assessments, their methods and findings are summarized here for reference.

2.2.1 Fuel Age Impacts on Gaseous Fission Product Capture during Separations

A report entitled Fuel Age Impacts on Gaseous Fission Product Capture during Separations (referred to within this report as the “Fuel Age Report”) was authored by Jubin et al. in 2012 (Jubin et al. 2012a). This report provided a summary of the three regulations that may be applied to a reprocessing facility (40 CFR 41, 40 CFR 190, and 10 CFR 20) and assessed how they might apply to an aqueous-based reprocessing facility with a throughput rate of 1,000 MTIHM/y. Three fuel types were used as source terms: pressurized water reactor (PWR) uranium oxide fuel, PWR mixed oxide fuel, and advanced high temperature gas-cooled reactor fuel. The analysis reviewed multiple burnups for each fuel type and estimated the required DFs for each fuel type processed as a function of cooling time following discharge from the nuclear reactor.

The applicable regulations include limits on both the total quantity of emissions of specific radionuclides (^{85}Kr, ^{129}I, and ^{239}Pu and other alpha-emitting TRU radionuclides with half-lives less than 1 y, referred to within this document as ^{239}Pu-TRU$_{<1y}$) and on the maximum dose that may be received by a member of the general public as a result of nuclear fuel cycle facility emissions. A conservative case in which volatile radionuclide emissions are limited to 10% of the dose to the maximum exposed individual (MEI) was considered in addition to a case in which 100% of the allowable dose was attributed to emitted volatile radionuclides. Distribution of iodine speciation between particulate and vapor phase was also examined, and it was found that assuming particulate iodine results in greater doses to the MEI and thus higher mitigation requirements.

The dose to the MEI was assessed using the EPA model Clean Air Act Assessment Package – 1988 (CAP-88) (CAP-88 PC 2020), which is a publicly available tool for air dispersion modeling. Key variables serving as input to the air dispersion modeling include the distribution of radionuclides between particulate and vapor species, the stack height and stack gas velocity of the facility, the geographical location and meteorological characteristics of that location, agricultural data, etc.

The results of the Fuel Age Report found that the mitigation requirements for the short-lived volatile radionuclides emitted from a 1,000 MTIHM/y aqueous-based reprocessing facility are highly dependent on the cooling time of the fuel being processed. In the processing of shorter-cooled (≤15 y) fuel, the maximum required DFs for ^{3}H and ^{85}Kr were estimated as 720 and 60, respectively. Mitigation for ^{14}C was not required in any but the most restrictive sets of cases. Iodine-129 mitigation requirements did not vary with fuel age, and the most restrictive potential DF for ^{129}I was 8,000. The dose-based DFs of both long-lived and short-lived radionuclides were found to depend on stack design and site-specific parameters.

2.2.2 Radioactive Semivolatiles in Nuclear Fuel Reprocessing

A report was issued in 2014 by Jubin and colleagues entitled Radioactive Semi-Volatiles in Nuclear Fuel Reprocessing (referred to within this report as the Semivolatiles Report) and was a follow-on effort to the Fuel Age Report. The objective of the Semivolatiles Report was to assess less-volatile radionuclides for their potential to contribute to the dose received by the MEI. Twenty-four semivolatile radionuclides present in uranium oxide UNF were identified for evaluation. Each radionuclide was evaluated based upon known chemical behavior, quantities present within the fuel, and with air dispersion modeling to determine their potential contribution to the dose received by the MEI. It was determined that there were eight radionuclides that could merit control based on their radioactivity/quantity within the fuel.

2.2.3 Separation and Waste Form Campaign Full Recycle Case Study

The issuance of Separation and Waste Form Campaign Full Recycle Case Study (referred to within this report as the Case Study Report) occurred in 2015 and was intended to develop reference sets of technologies and preliminary mass balances for both aqueous and electrochemical reprocessing technologies (Law et al. 2015). The reference case analyzed was the aqueous reprocessing of uranium oxide fuel to recover fuel for an SFR. The used fuel discharge from the SFR was then to be electrochemically reprocessed for recovery of fissile material and fuel refabrication. Importantly, the Case Study Report extended its detailed mass balance and technology assessments to waste treatment associated with both aqueous and electrochemical reprocessing. Included within the waste treatment flowsheets were conceptual designs for off-gas management based on the requirements outlined in the Fuel Age Report. For electrochemical reprocessing, the systems were broadly aligned with the design of the IFR reprocessing operations. The reference system described in the Case Study Report, the associated UNF source term, and the developed mass balances were all used as resources in the development of this report.

2.3 Applicability of Prior Work

In reviewing prior facility designs, emissions assessments, and potential mitigation requirements, several key differences between previous work and the scenario evaluated in this report were identified.

First, as aqueous reprocessing has been implemented at an industrial scale, available design information and process knowledge are significantly more detailed than corresponding information for electrochemical reprocessing. As a result, assessments of aqueous reprocessing can be performed with more granularity than assessments of electrochemical reprocessing. Key facility design information that would affect off-gas system design (such as likely cell gas purification processes, likely stack design, and specific processing parameters, including temperature, batch size, etc.) is still conceptual in nature for electrochemical reprocessing. As a result, the assessment completed in this report must have some level of unavoidable imprecision arising from the evaluation of a "generic" facility with less defined design and operational specifics.

Second, the expected throughput of an industrial-scale electrochemical reprocessing facility is expected to be substantially lower than an industrial-scale aqueous reprocessing facility. This characteristic, along with the expectation that fuel will be cooled <5 y before electrochemical reprocessing, results in differences in the quantity of radioisotopes present in the fuel to be processed. Cooling time is especially influential (as revealed in the Fuel Age Report) as additional decay time can significantly lower mitigation requirements for ^{3}H and ^{85}Kr.

Finally, the selection of metallic fuel as compared to oxide-based fuel is important. A key driver in the ^{14}C source term is the presence of nitrogen impurities within oxide fuel. These nitrogen impurities will activate within a nuclear reactor and the activation and subsequent decay to ^{14}C is effectively the primary source term for ^{14}C in UNF. Metallic fuel is not expected to contain measurable levels of nitrogen impurities, effectively eliminating ^{14}C as a concern in the reprocessing of metallic fuel. Note, some SFRs are designed to use oxide fuel, and in this case the electrochemical processing of this fuel would have to account for potential ^{14}C release requirements.

Table 1 provides a comparison of volatile radionuclide quantities expected in uranium oxide-based PWR fuel (irradiated to 60 GWd/MTIHM and cooled for 5 y) and in metallic alloy uranium fuel (irradiated to 99.9 GWd/MTIHM and cooled for 2 y). The information included in this table was sourced from the Fuel Age Report and the Case Study Report. With the exception of ^{14}C, the quantities of volatile radionuclides produced are within an order of magnitude of each other. However, understanding that the likely throughput of an electrochemical reprocessing facility could be ≤20% of an aqueous facility, the source term for assessment of a conceptual electrochemical reprocessing facility will decrease accordingly.

Table 1: Predicted radioisotope quantities present in UNF discharged from a reference case PWR and a reference case SFR.

Radionuclide	Specific Activity	g/MTIHM		Ci/MTIHM	
	Ci/g	PWR	SFR	PWR	SFR
^{3}H	9,800	1.39E-01	2.28E-01	1.36E+03	2.23E+03
^{14}C	4.46	3.76E-01	0	1.68E+00	0.00E+00
^{85}Kr	400	3.51E+01	3.87E+01	1.40E+04	1.55E+04
^{129}I	0.00018	2.91E+02	7.55E+02	5.24E-02	1.36E-01

Note: The PWR reference case is uranium oxide fuel irradiated to a burnup of 60 GWd/MTIHM and cooled for 5 y after reactor discharge. The SFR reference case is metallic uranium fuel irradiated to a burnup of 99.6 GWd/MTIHM and cooled for 2 y after reactor discharge.

2.4 Regulations Applicable to Evaluated Scenario

A full examination of the applicable regulations is provided in Jubin et al. (2012a). Here, key aspects regarding the three primary regulations are briefly summarized for reference.

Volatile radionuclide emissions from a nuclear fuel recycle facility are addressed in several regulatory documents. The EPA has established, through 40 CFR 190, annual dose limits for nuclear fuel cycle facilities in the commercial sector (EPA 2010b). In 40 CFR 190.10(a), the dose limits for specific organs and for the whole body are provided. Specific release limits for ^{85}Kr, ^{129}I, and ^{239}Pu-TRU$_{<1y}$ in curies (Ci) released per unit of electric power produced are also defined in 40 CFR 190.10(b) (EPA 2010b). Dose limits for workers and individual members of the public for facilities have been established in 10 CFR 20 for activities conducted under licenses issued by the NRC (NRC 2012). Dose limits at US DOE facilities for workers and individual members have been established in 40 CFR 61.92 (EPA 2010a).

2.4.1 40 CFR 190

The provisions of this section are applicable to radiation doses received by members of the public in the general environment and to radioactive materials introduced into the general environment as the result of operations that are part of a nuclear fuel cycle. Section 40 CFR 190.10 provides the standards for normal operation and is notable in that it prescribes limits to the total annual dose to any member of the public and also prescribes specific release limits for ^{85}Kr, ^{129}I, and ^{239}Pu-TRU$_{<1y}$:

> *§ 190.10(a): 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

> *§ 190.10(b): 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.*

Depending on the used fuel compositions, specific radionuclides, the reprocessing facility design, reprocessing rate, and stack gas air dispersion parameters, either the dose-based [40 CFR 190.10(a)] or fuel cycle-based [40 CFR 190.10(b)] limits may impose more restrictive radionuclide emissions control for a reprocessing facility.

2.4.2 10 CFR 20

This regulation establishes “standards for protection against ionizing radiation resulting from activities conducted under licenses issued by the Nuclear Regulatory Commission. These regulations are issued under the Atomic Energy Act of 1954, as amended, and the Energy Reorganization Act of 1974, as amended” (NRC 2012).

Sections of 10 CFR 20 that apply to this analysis of compliance to dose regulations for the gaseous fission products are excerpted below for reference. Unlike 40 CFR 190.10(b), specific release limits are not prescribed; rather the total dose arising from an NRC-licensed facility to an exposed member of the public (excluding occupational exposure and other explicitly excluded conditions) must fall below 100 mrem/y.

Section 10 CFR 20.1301 establishes the dose limits for individual members of the public and states the following:

> *(a) Each licensee shall conduct operations so that—*
>
> > *(1) The total effective dose equivalent to individual members of the public from the licensed operation does not exceed 0.1 rem (1 mSv) in a year, exclusive of the dose contributions from background radiation, from any medical administration the individual has received, from exposure to individuals administered radioactive material and released under § 35.75, from voluntary participation in medical research programs, and from the licensee’s disposal of radioactive material into sanitary sewerage in accordance with § 20.2003, and*
> >
> > *(2) The dose in any unrestricted area from external sources, exclusive of the dose contributions from patients administered radioactive material and released in accordance with § 35.75, does not exceed 0.002 rem (0.02 mSv) in any one hour.*

> *(b) In addition to the requirements of this part, a licensee subject to the provisions of EPA's generally applicable environmental radiation standards in 40 CFR part 190 shall comply with those standards.*

Demonstration of compliance to 10 CFR 20.1301 is expected to proceed as described in 10 CFR 20.1302, with key excerpts provided here:

> *(a) The licensee shall make or cause to be made, as appropriate, surveys of radiation levels in unrestricted and controlled areas and radioactive materials in effluents released to unrestricted and controlled areas to demonstrate compliance with the dose limits for individual members of the public in § 20.1301.*
>
> *(b) A licensee shall show compliance with the annual dose limit in § 20.1301 by—*
>
> > *(1) Demonstrating by measurement or calculation that the total effective dose equivalent to the individual likely to receive the highest dose from the licensed operation does not exceed the annual dose limit; or*
> >
> > *(2) Demonstrating that—*
> >
> > > (i) *The annual average concentrations of radioactive material released in gaseous and liquid effluents at the boundary of the unrestricted area do not exceed the values specified in Table 2 of Appendix B to Part 20; and*
> > >
> > > (ii) *If an individual were continuously present in an unrestricted area, the dose from external sources would not exceed 0.002 rem (0.02 mSv) in an hour and 0.05 rem (0.5 mSv) in a year.*
>
> *(c) Upon approval from the Commission, the licensee may adjust the effluent concentration values in appendix B to part 20, table 2, for members of the public, to take into account the actual physical and chemical characteristics of the effluents (e.g., aerosol size distribution, solubility, density, radioactive decay equilibrium, chemical form).*

For the purposes of this study, we do not consider the short-term limit of 0.002 rem (0.02 mSv) in 20.1302(b)(2)(ii) because the lower annual limit is more restrictive.

10 CFR 20 Appendix B defines limits for the annual average concentration of radioactive material that can be present at the site boundary of a UNF reprocessing facility. These limits are derived by allocating a 50 mrem/y dose to an individual maximally exposed member of the public.

2.4.3 40 CFR 61

This part applies to operations at any facility owned or operated by DOE from which any radionuclide other than ^{222}Rn and ^{220}Rn is emitted into the air, except that this part does not apply to disposal at facilities subject to 40 CFR part 191 subpart B or 40 CFR part 192 (EPA 2010a).

This regulation establishes that the

> *emissions of radionuclides to the ambient air from Department of Energy facilities shall not exceed those amounts that would cause any member of the public to receive in any year an effective dose equivalent of 10 mrem/yr.*

It is also noted that

> *demonstration of compliance will be based on 10 CFR 20 methodology by reduction of DAC*[a] *by appropriate factor of 5 (see notes on development of Table 2 (Appendix B to Part 20).*

40 CFR 61 is thus more restrictive than both 40 CFR 190 and 10 CFR 20, with specific implementation and compliance aspects mimicking 10 CFR 20.

2.4.4 Assumptions Regarding Regulatory Compliance

This study assumes that the facility examined is commercially operated and licensed under the NRC and is not a DOE facility, thus eliminating the more restrictive 40 CFR 61 requirements.

The 0.1 rem/y (100 mrem/y) dose limit in 10 CFR 20.1301(a)(1) and the 50 mrem/y dose that is the basis for the concentration limits at the site boundary in 10 CFR 20 Appendix B are both less restrictive than the dose limit of 25 mrem/y in 40 CFR 190.10. As a result, for the purposes of the analyses described by this report, compliance to 40 CFR 190.10 also ensures compliance with 10 CFR 20.

Therefore, the analyses described by this report are based primarily on compliance with both dose- and fuel cycle-based 40 CFR 190.10 limits for radionuclide emissions.

Additional assumptions are listed here:

1. No engineering margins will be applied.
2. The site boundary will not prevent public access to the location of the MEI as determined by CAP-88.
3. The potential for release of ^{85}Kr or ^{129}I, and ^{239}Pu-TRU$_{<1y}$ in other parts of the fuel cycle is not considered. If discrete amounts of these radionuclides are released to the atmosphere in other parts of a fuel cycle (such as during reactor operations or during used fuel storage before reprocessing), then the control efficiencies estimated in this study for compliance to the fuel cycle limits in 40 CFR 190.10(b) would need to be proportionately higher.
4. The reprocessing facility is not co-located with any other nuclear facilities, such that they would be considered a single facility, which would require DFs calculated based on dose to be based on total radionuclide emissions from all co-located nuclear facilities.

To meet these release restrictions for a plant, certain DFs are needed for the removal of these radionuclides from the gaseous effluent. Discussion of likely DF requirements and their associated bases is provided in Section 4.

3. DESCRIPTION OF REFERENCE CASE USED FUEL

The irradiated metallic fuel chosen as the source term for the assessment described in this report is illustrated in Figure 2. This is an example prototype Generation IV SFR metallic driver fuel. Blanket fuels are also used in SFRs, but only the driver fuel is assessed in this report because driver fuels typically experience higher burnup and correspondingly higher levels of activation and fission products. The reference fuel assembly has a hexagonal stainless-steel duct that encloses a matrix of fuel rods or pins and other stainless-steel hardware that does not contact the actual fuel.

---

[a] The concentration of a given radionuclide in air which, if breathed by the reference man for a working year of 2,000 hours under conditions of light work (with an inhalation rate of 1.2 cubic meters of air per hour), results in an intake of one annual limit on intake.

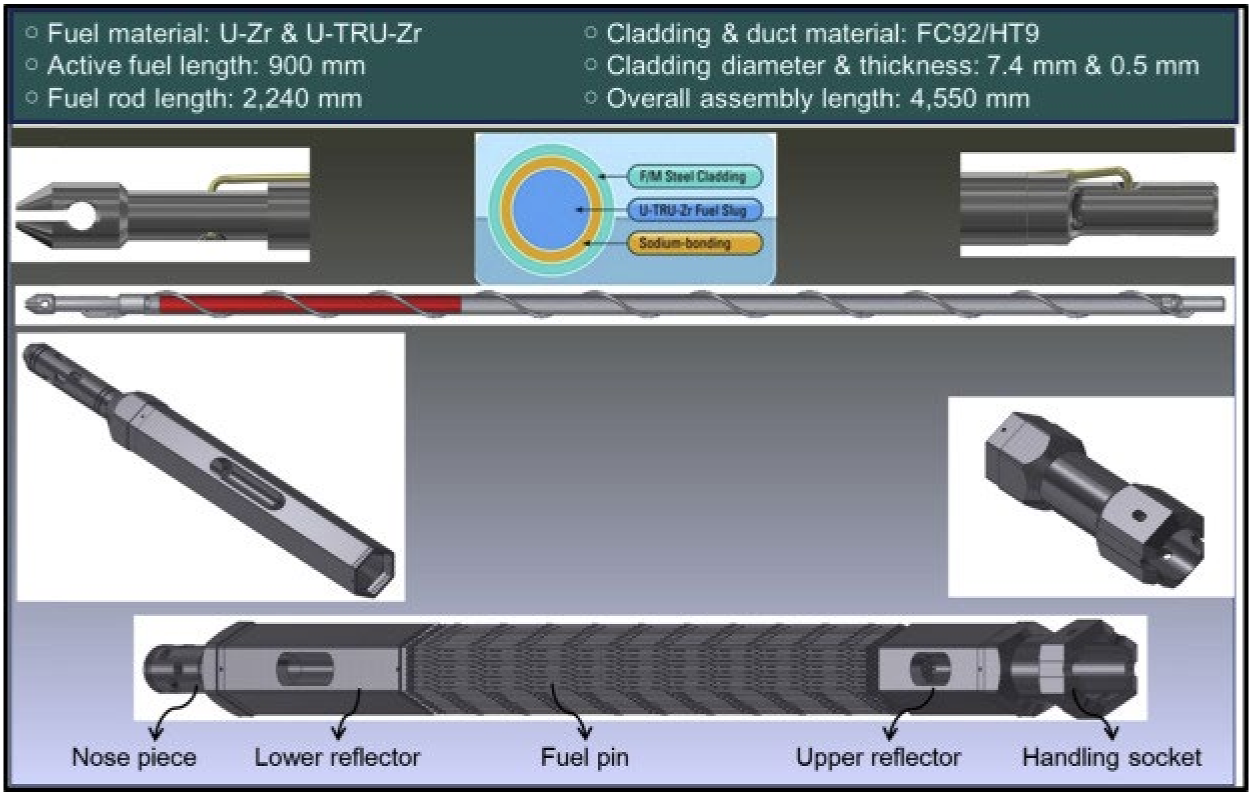

Figure 2: Example prototype Generation IV SFR metal fuel assembly (from Lee 2016).

Figure 3 illustrates a fuel-bearing segment of a fuel pin with stainless-steel cladding that, before irradiation, contains the fuel slugs with the bond sodium. The bond sodium provides a solid bond for efficient heat transfer between the fuel slugs and the cladding and molten sodium reactor coolant that flows outside of the fuel pins. The gas plenum provides volume for fission product gases, including Kr and Xe, to expand into during irradiation. After sufficient irradiation in the reactor, the fuel swells until it can contact the cladding and displace the (molten) bond sodium into the plenum along with fission product gases. Other fission products including Cs and I can also migrate into the plenum. These details are relevant in electrorefining because the plenum, along with the pin that contains fuel, is chopped for processing in the ER to recover the reusable actinides and separate the fission products for disposal.

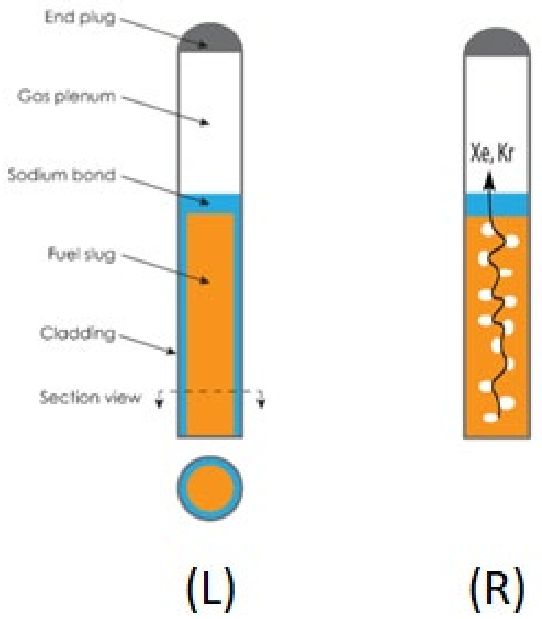

Figure 3: Illustration of a fuel pin segment showing the gas plenum above the fuel section inside the stainless-steel cladding before *(left)* and after *(right)* irradiation (from FRWG 2018).

The metallic uranium fuel selected as the source term is assumed to be alloyed with zirconium and plutonium and to be at equilibrium within a closed fuel cycle with a conversion ratio of 0.75. Initial fuel composition is assumed to be 70wt%U–20 wt% TRU–10 wt% Zr. The equilibrium composition of the TRU components of the used fuel for the system with a conversion ratio of 0.75 is 0.21% Np–17.7 wt% Pu–0.92 wt% Am–0.38 wt% Cm.

The composition of this example used fuel shown in Table 2 was calculated using the ORIGEN model. This composition does not include any cladding, plenum, duct, or other non-fuel masses.

Table 2: SFR used metal fuel (99.6 GWd/MTIHM, 2-y cooled, 0.75 conversion ratio) (Law et al. 2015).

Concentration ***(g/MTIHM)***								
Total[a]								
1.13E+06								
Total H	H 3[b]							
2.28E-01	2.28E-01							
Total C	C 12	C 14						
1.75E+02	1.75E+02	0.00E+00						
Total Na[c]								
2.40E+04								
Total Se	SE 76	SE 77	SE 78	SE 79	SE 80	SE 82		
9.59E+01	3.85E-02	3.18E+00	6.83E+00	1.20E+01	1.98E+01	5.41E+01		
Total Kr	KR 80	KR 81	KR 82	KR 83	KR 84	KR 85	KR 86	
6.88E+02	9.25E-04	4.06E-05	2.04E+00	1.15E+02	2.09E+02	3.87E+01	3.23E+02	
Total Rb	RB 85	RB 87						
6.13E+02	1.89E+02	4.25E+02						
Total Sr	SR 86	SR 87	SR 88	SR 89	SR 90			
1.33E+03	7.19E+00	1.42E-01	5.53E+02	1.34E-03	7.74E+02			
Total Y	Y 89	Y 90	Y 91					
7.26E+02	7.26E+02	1.94E-01	8.52E-03					
Total Zr[d]	ZR 90	ZR 91	ZR 92	ZR 93	ZR 94	ZR 95	ZR 96	
1.19E+05	9.38E+01	1.13E+05	1.22E+03	1.50E+03	1.73E+03	3.70E-02	1.99E+03	
Total Nb	NB 93	NB 93M	NB 94	NB 95	NB 95M			
5.58E-02	5.60E-04	2.68E-03	7.96E-03	4.46E-02	2.66E-05			
Total Mo	MO 95	MO 96	MO 97	MO 98	MO100			
9.07E+03	1.87E+03	5.72E+01	2.09E+03	2.35E+03	2.69E+03			
Total Tc	TC 98	TC 99						
2.39E+03	0.00E+00	2.39E+03						
Total Ru	RU 99	RU100	RU101	RU102	RU103	RU104	RU106	
8.62E+03	3.28E-02	1.05E+02	2.53E+03	3.07E+03	2.74E-04	2.77E+03	1.43E+02	
Total Rh	RH102	RH103	RH103M	RH106				
2.96E+03	0.00E+00	2.96E+03	2.70E-07	1.34E-04				
Total Pd	PD104	PD105	PD106	PD107	PD108	PD110		
6.45E+03	1.73E-02	2.03E+03	1.94E+03	1.20E+03	1.01E+03	2.75E+02		
Total Ag	AG107	AG108	AG108M	AG109	AG109M	AG110	AG110M	
6.45E+02	5.50E-04	3.59E-14	1.32E-05	6.45E+02	9.70E-14	1.28E-09	8.09E-02	
Total Cd	CD108	CD109	CD110	CD111	CD112	CD113	CD113M	CD114
4.32E+02	2.62E-05	9.60E-08	6.01E+01	1.43E+02	9.95E+01	5.10E+01	2.92E+00	4.71E+01
	CD115M	CD116						
	2.21E-06	2.83E+01						

Table 2, (continued): SFR used metal fuel (99.6 GWd/MTIHM, 2-y cooled, 0.75 conversion ratio) (Law et al. 2015).

Concentration ***(g/MTIHM)***								
Total In	IN113	IN113M	IN114	IN114M	IN115			
3.37E+01	5.70E-01	0.00E+00	8.15E-13	5.03E-08	3.32E+01			
Total Sn 3.35E+02	SN114	SN115	SN116	SN117	SN118	SN119	SN119M	SN120
	1.50E-02	1.11E+00	2.48E+00	2.78E+01	2.98E+01	2.84E+01	2.86E-02	2.96E+01
	SN121M	SN122	SN123	SN124	SN126			
	6.13E-02	3.33E+01	4.29E-02	5.29E+01	1.30E+02			
Total Sb	SB121	SB123	SB124	SB125	SB126	SB126M		
1.02E+02	3.03E+01	3.96E+01	3.04E-05	3.16E+01	6.16E-06	4.69E-08		
Total Te 1.71E+03	TE122	TE123	TE123M	TE124	TE125	TE125M	TE126	TE127
	1.20E+00	1.73E-02	2.14E-05	1.40E+00	5.63E+01	4.43E-01	7.29E+00	1.30E-04
	TE127M	TE128	TE129	TE129M	TE130			
	3.71E-02	4.12E+02	1.24E-09	1.34E-06	1.23E+03			
Total I	I127	I129						
1.00E+03	2.49E+02	7.53E+02						
Total Xe	XE128	XE129	XE130	XE131	XE132	XE134	XE136	
1.27E+04	1.47E+01	1.03E-01	1.35E+00	2.08E+03	2.98E+03	3.99E+03	3.68E+03	
Total Cs	CS133	CS134	CS135	CS137				
1.12E+04	3.61E+03	4.67E+01	4.08E+03	3.42E+03				
Total Ba	BA134	BA135	BA136	BA137	BA137M	BA138		
4.32E+03	9.95E+01	7.20E-02	1.78E+02	3.59E+02	5.21E-04	3.69E+03		
Total La	LA138	LA139						
3.39E+03	5.11E-03	3.39E+03						
Total Ce	CE140	CE142	CE144					
6.09E+03	3.19E+03	2.81E+03	8.85E+01					
Total Pr	PR141	PR144	PR144M					
3.26E+03	3.26E+03	3.74E-03	1.87E-05					
Total Nd	ND142	ND143	ND144	ND145	ND146	ND148	ND150	
1.02E+04	2.89E+01	2.57E+03	2.35E+03	1.77E+03	1.78E+03	1.07E+03	6.47E+02	
Total Pm	PM147	PM148	PM148M					
4.10E+02	4.10E+02	1.67E-07	1.86E-05					
Total Sm	SM146	SM147	SM148	SM149	SM150	SM151	SM152	SM154
2.99E+03	0.00E+00	7.79E+02	1.74E+02	7.96E+02	1.11E+02	4.53E+02	4.82E+02	1.92E+02
Total Eu	EU150	EU151	EU152	EU153	EU154	EU155		
3.46E+02	0.00E+00	1.38E+01	5.29E-01	2.28E+02	2.54E+01	7.85E+01		
Total Gd	GD152	GD153	GD154	GD155	GD156	GD157	GD158	GD160
3.02E+02	3.98E-01	3.14E-04	8.82E+00	5.98E+01	1.08E+02	5.81E+01	5.24E+01	1.44E+01
Total Tb	TB159	TB160						
2.21E+01	2.21E+01	4.01E-04						

Table 2, (continued): SFR used metal fuel (99.6 GWd/MTIHM, 2-y cooled, 0.75 conversion ratio) (Law et al. 2015).

Concentration *(g/MTIHM)*								
Total Ho	HO165	HO166M						
5.10E-01	5.09E-01	9.05E-04						
Total Tl	TL207	TL208	TL209					
7.28E-10	1.33E-14	7.28E-10	2.06E-16					
Total Pb	PB206	PB207	PB208	PB209	PB210	PB211	PB212	PB214
5.95E-04	9.50E-12	2.02E-09	5.95E-04	8.44E-13	2.28E-10	1.02E-13	4.23E-07	8.04E-15
Total Bi	BI209	BI210	BI211	BI212	BI213	BI214		
1.69E-07	1.28E-07	1.49E-13	6.10E-15	4.03E-08	2.04E-13	5.91E-15		
Total Po	PO210	PO216	PO218					
4.92E-12	3.23E-12	1.69E-12	9.32E-16					
Total Rn	RN219	RN220	RN222					
6.46E-10	1.96E-16	6.44E-10	1.71E-12					
Total Fr	FR221	FR223						
2.26E-14	2.16E-14	9.20E-16						
Total Ra	RA223	RA224	RA225	RA226	RA228			
3.95E-06	4.92E-11	3.68E-06	9.78E-11	2.67E-07	9.32E-15			
Total Ac	AC225	AC227	AC228					
3.46E-08	6.61E-11	3.45E-08	9.72E-19					
Total Th	TH227	TH228	TH229	TH230	TH231	TH232	TH234	
1.04E-02	7.88E-11	7.15E-04	1.79E-05	9.58E-03	1.07E-09	7.25E-05	1.01E-05	
Total Pa	PA231	PA233	PA234	PA234M				
5.50E-04	4.79E-04	7.09E-05	1.17E-10	3.39E-10				
Total U	U232	U233	U234	U235	U236	U237	U238	U240
7.00E+05	4.49E-02	2.12E-02	6.38E+02	2.64E+02	4.15E+02	2.42E-04	6.99E+05	1.38E-12
Total Np	NP237	NP239	NP240M					
2.06E+03	2.06E+03	3.33E-03	1.19E-14					
Total Pu	PU236	PU238	PU239	PU240	PU241	PU242	PU243	PU244
1.77E+05	2.47E-02	4.85E+03	9.39E+04	5.79E+04	8.08E+03	1.23E+04	1.06E-09	7.21E-02
Total Am	AM241	AM242	AM242M	AM243	AM244			
9.25E+03	4.93E+03	3.48E-03	2.90E+02	4.03E+03	5.60E-17			
Total Cm	CM242	CM243	CM244	CM245	CM246	CM247	CM248	
3.79E+03	1.13E+01	1.93E+01	2.61E+03	7.21E+02	3.83E+02	3.10E+01	1.43E+01	
Total Bk	BK249							
5.80E-02	5.80E-02							
Total Cf	CF250	CF251	CF252					
1.39E+00	1.75E-01	2.34E-02	8.73E-04					
a	*Does not include cladding and other non-fuel-bearing components.*							
b	*70% of this H-3 is assumed lost to reactor coolant.*							
c	*This is bond sodium.*							
d	*Includes both fission product Zr and the Zr alloy as Zr-91.*							

4. ESTIMATED DECONTAMINATION FACTORS NEEDED FOR REGULATORY COMPLIANCE

Overall facility DFs needed to meet air emission limits can be calculated using an assumed input used fuel burnup and composition, assumed electrochemical processing annual throughput rate, and air dispersion modeling to calculate dose maps for the surrounding area, which in turn leads to the maximum dose equivalent to the public. The overall facility DF is ratio of the maximum uncontrolled radionuclide emission rate (or dose caused by the maximum uncontrolled radionuclide emission rate) divided by the regulatory limits. If the electrochemical process can inherently achieve the needed DFs because of its process design and operation, no additional off-gas emissions control is needed. But if not, the needed DFs must be achieved either by process design, operating modifications, or by adding off-gas control.

Using assumptions detailed in Section 2 and the fuel composition provided in Section 3, the facility-wide DFs needed for compliance to both 40 CFR 190.10(a) and 40 CFR 190.10(b) were determined assuming total uncontrolled release of key radionuclides from the incoming fuel. Air dispersion modeling was used to develop dose maps surrounding the stack of a facility and determine the dose equivalent to the maximum exposed individual (MEI). The DFs needed to comply with the dose equivalent limit in 40 CFR 190.10(a) were determined by dividing the calculated dose equivalent by the regulatory dose equivalent limit. The needed DFs for compliance with 40 CFR 190.10(b) for ^{85}Kr, ^{129}I, and ^{239}Pu-TRU$_{<1y}$ are the maximum uncontrolled emission rates in curies (Ci) or millicuries per gigawatt per year (mCi/GWy) divided by the regulatory limits.

The regulatory emission limits and dose limits for individual radionuclides that are in the groupings of (a) ^{239}Pu-TRU$_{<1y}$ and (b) individual radionuclides that contribute to the total dose calculation need to be prorated so that the total emission rate or dose for the group is within the regulatory limits for the entire group. The approach used in this report when calculating needed DFs for each radionuclide in a group of radionuclides is to limit the allowable emission rate or dose for each radionuclide in a group to 10% of the regulatory limit for the group. This approach is a simple way to make each single radionuclide a small contributor to the total emission rate or dose from the group. If the emission rate or dose for each radionuclide in the group is less than 10% of the regulatory limit for the group, then the total emission rate or dose for the group can be expected to meet the group regulatory limit. This 10% approach has precedence in other areas such as in the determination of Class A versus Class C low-level waste and in State of Idaho determinations of “Below Regulatory Concern” for air pollutant emission rates that are 10% or less of the significant emission rates (NRC 2001, Idaho Department of Environmental Quality 2020).

Using the 10% approach outlined above, the dose limit for each radionuclide likely to contribute to dose to the MEI is 2.5 mrem/y. Elements that could be in used nuclear fuels that are most commonly assumed to be volatile during reprocessing include the noble gas krypton and other elements that might exist in the form of gaseous species at ambient temperature and pressure, or under reprocessing conditions, such as tritium (in the form of diatomic tritium or tritiated water) and iodine (in the form of diatomic iodine or other iodides). As described above, alpha-emitting TRU isotopes are also considered in dose modeling, with the understanding that most are refractory in nature and so can be expected to be in particulate form in process gas streams. Dose limits are determined through CAP-88 modeling described in sections below.

Concentrations of ^{239}Pu-TRU$_{<1y}$ in the fuel are listed in Table 3. Of the nuclides listed in this table, only ^{239}Pu and the three radionuclides with the highest activities within the fuel (^{238}Pu, ^{241}Am, and ^{244}Cm) have been included in the air dispersion modeling.

Table 3: Activities in the used fuel of alpha-emitting TRU with half-lives greater than 1 y.

TRU isotopes	Specific Activity (Ci/g)	$t_{1/2}$ [a] (y)	Activity, Ci/MTIHM
Am-241[b]	**3.43E+00**	**433**	**1.58E+04**
Cf-252	5.36E+02	3	4.71E-01
Cm-244[b]	**8.09E+01**	**18**	**2.14E+05**
Cm-245	1.72E-01	8,500	1.23E+02
Cm-248	4.14E-03	348,000	6.15E-02
Np-237	7.05E-04	2,140,000	1.42E+00
Pu-238[b]	**1.71E+01**	**88**	**8.29E+04**
Pu-239[b]	**6.21E-02**	**24,100**	**5.76E+03**
Pu-240	2.27E-01	6,560	1.31E+04
Pu-242	3.94E-03	375,000	4.83E+01
Pu-244	1.83E-05	80,000,000	1.36E-06
		Total ^{239}Pu/TRU activity *(Ci/MTIHM)*	3.30E+05

[a] $t_{1/2}$ sourced from Baum et al. 2002.

[b] Bolded isotopes were selected for inclusion in air dispersion modeling for dose calculation; ^{239}Pu because it is specifically identified in the regulation, and the other three because they represent the highest activity of the alpha-emitting TRU with half-lives greater than 1 y within the fuel.

4.1 Emissions Limits Based on 40 CFR 190.10(a)

The facility DFs required to comply with 40 CFR 190.10(a) were estimated using air dispersion modeling. CAP-88 is a set of computer programs, databases, and associated utility programs used to estimate dose and risk to members of the public from radionuclide emissions in the air. Version 4.1 of CAP-88 incorporates dose and risk factors from Federal Guidance Report 13 (EPA 1999), which are based on the methods of the International Commission on Radiological Protection (ICRP 1996). Emission monitoring and compliance procedures for DOE facilities require the use of the CAP-88 model or other approved methodologies to estimate the effective dose to members of the public.

4.1.1 Use of CAP-88 Model

The three main pathways for exposure from an atmospheric release of radiological material considered in CAP-88 are ingestion, inhalation, and external exposure. Ingestion is from consumption of plants, animals, fish, or water contaminated with radionuclides. Inhalation occurs when a person is in the pathway of a gas plume containing radioactive materials. External exposure occurs for people who live or recreate in areas where the ground or water has been exposed to radiological materials. Assessments for collective populations or an MEI are possible. Dose and risk estimates from CAP-88 are applicable only to low-level chronic exposures because the health effects and dosimetric data are based on low-level chronic intakes. The model cannot be used for either short-term or high-level radionuclide intakes.

To use the CAP-88 model, various location-specific, design-specific, and operational parameters are required as input for the simulation. The following were used to form the base case:

1. **Facility Data**—This input is for general descriptive information of the facility; however, the only information used in calculations is the state in which the facility is located. The state parameter determines the appropriate agricultural data (cattle and crop production) to be used in the simulation. Data are available for all states; however, in this study we focus on a hypothetical facility located in Idaho or Tennessee to mimic two disparate climates within the US.

2. **Run Options**—This input is for an individual or a collective population. The individual option was used in this study so the results could be applied to an assumed MEI.

3. **Meteorological Data**—The local average weather conditions are required and include wind characteristics (16 vector description), annual rainfall, average ambient temperature, humidity, and meteorological lid. Calculations performed for each site used meteorological data in the CAP-88 model considered representative of the site. The additional meteorological data necessary for these simulations can be seen below in Table 4. In this study we have used annual averages, which reflect the regulatory language regarding the annual dose to the MEI.

4. **Source Data (Facility Design and Operation)**—This input is for the source of the emission. A stack release is assumed for a reprocessing plant, and the data required includes the stack dimensions. The plume type is also required, and choices include buoyant or momentum and Pasquill stability class categories for plumes rising above the stack. In this sensitivity study only a momentum plume type was considered. The values used for the base case are a stack height of 50 m (164 ft); a stack diameter of 2.2 m; and a momentum-type plume with a stack exit velocity of 12.2 m/s (corresponding to a volumetric rate of 46 m^3/s or 98,000 scfm). This reference case was selected to be consistent with prior air dispersion studies, including Soelberg 2008.

5. **Agricultural Data**—This input is used to estimate the uptake of nuclides into the food chain based on agricultural use in the area. Choices are urban, rural, local, regional, or imported. Although values are automatically selected based on the state (location) of the source, the agricultural data can be

defined by the user if desired. The local case was assumed for this study as it results in the worst case in terms of ingestion dose.

6. **Radionuclide Data**—This input identifies the nuclides emitted from the source, their emission rates in curies per year, the physical form for some radionuclides (particle, vapor, or organic), and chemical form for some species (tritium). The vapor form of ^{3}H in tritiated water was assumed; ^{85}Kr form was an unspecified gas; ^{129}I form was vapor but a sensitivity study performed using both particulate and vapor is described in Section 4.1.3. Modeling of the TRU nuclides was performed for ^{239}Pu, ^{241}Am, ^{238}Pu, and ^{244}Cm as particulate.

Table 4: Meteorological data used in CAP-88 simulations.

Location	Annual precipitation (cm/y)	Annual ambient temperature (°C)	Lid height[a] (m)	Absolute humidity (g/m^{3})	Data source
Pocatello, ID	29.44	8.15	1000	4.8	Western Regional Climate Center
Knoxville, TN	121.56	15.07	1000	10.3	NOAA

[a] Lid height refers to the average height of the tropospheric mixing layer (or atmospheric boundary layer) at the site. Jubin et al. (2012) showed minimal effects of varied lid height on dose to the MEI.

4.1.2 Baseline Dose Modeling for Facilities in Idaho and Tennessee

Climatology and topography, determined by facility location, can affect the location of and dose to the MEI. In this study we investigate a hypothetical electrochemical reprocessing facility potentially located in Idaho and Tennessee. These two hypothetical locations have been selected as base cases to generally encompass the range of climatological variation that can be found in the US.

CAP-88 uses algorithms within the program to calculate the dose at radial coordinates from the stack location. The program then determines the single highest dose from this map and identifies this as the MEI. In this report we will present a selection of the dose maps generated from the radial coordinate data to illustrate the complexity of the stack release in the environment, as well as specific tabulated MEI dose data used in the calculation of DFs.

Tables 5 and 6 show the DFs and the location of the MEI for the uncontrolled release of the isotopes studied from a 200 MTIHM/y throughput facility with a 50 m stack that has a 2.2 m stack diameter releasing at a velocity of 12.2 m/s located in Idaho and Tennessee, respectively. Dose as a function of distance from the stack and the location of the MEI is determined by CAP-88 as the position of the highest single effective dose equivalent, and the DF is calculated from this MEI effective dose equivalent.

Table 5: Summary of required DFs for radioisotopes to meet EPA dose regulations for a facility located in Idaho. The maximum DF (resulting from either the whole body or thyroid dose) is bolded.

Isotope	Whole Body DF[a] (10%)	Thyroid DF (10%)	Location of MEI
Pu-238	**1.4E+08**	2.8E+06	900 m ENE
Pu-239	**2.8E+01**	1.4E-01	900 m ENE
Am-241	**1.3E+03**	3.3E+01	900 m ENE
Cm-244	**2.0E+08**	3.9E+06	900 m ENE
H-3	**1.1E+01**	3.5E+00	900 m ENE
Kr-85	**4.1E-01**	6.7E-02	900 m ENE
I-129 (vapor)	2.4E-01	**1.6E+00**	900 m ENE

[a] The whole body DF is based on a limit for each isotope of 2.5 mrem/y, 10% of the 25 mrem/y whole body dose limit. The thyroid DF is based on a limit of 7.5 mrem/y, 10% of the 75 mrem/y thyroid dose limit

Table 6: Summary of required DFs for radioisotopes to meet EPA dose regulations for a facility located in Tennessee. The maximum DF (resulting from either the whole body or thyroid dose) is bolded.

Isotope	Whole Body DF[a] (10%)	Thyroid DF (10%)	Location of MEI
Pu-238	**1.8E+08**	3.6E+06	500 m S
Pu-239	**3.5E+01**	6.9E-01	500 m S
Am-241	**1.6E+03**	4.6E+01	500 m S
Cm-244	**2.5E+08**	4.9E+06	500 m S
H-3	**6.9E+00**	2.2E+00	700 m NE
Kr-85	**5.0E-01**	8.2E-02	700 m NE
I-129 (vapor)	3.0E-01	**2.0E+00**	700 m NE

[a] The whole body DF is based on a limit for each isotope of 2.5 mrem/y, 10% of the 25 mrem/y whole body dose limit. The thyroid DF is based on a limit of 7.5 mrem/y, 10% of the 75 mrem/y thyroid dose limit

These tables show the direction and distance of the MEI varies depending on the location of facility, the specific isotope modeled, and the chemical form of the isotope. For the Idaho facility in this study, all particulate matter and vapor/gas result in an MEI that would be located 900 m ENE from the stack site.

The TRU isotopes (which are modeled as particulate phases in the CAP-88 model) in Tennessee have an MEI that is generally closer to the stack than the constituents considered as a gas or vapor. Note, the MEI resulting from ^{239}Pu-TRU$_{<1y}$ doses is located in a different direction (due south). The vapor or gases emitted from this facility results in an MEI that is 700 m in the NE direction, a 135° difference.

Broadly speaking, the dose-based DFs for tritium are on the order of 10. The dose-based DFs for ^{85}Kr are less than 1. The dose-based limits ^{129}I (vapor) are on the order of 2. The dose-based DFs for the ^{239}Pu-TRU$_{<1y}$ are observed to become as high as 10^8. The calculated dose-based DFs for ^{238}Pu and ^{244}Cm dominate by several orders of magnitude other ^{239}Pu-TRU$_{<1y}$ isotopes that have lower activities in the used fuel and lower doses to the MEI.

Across all cases and locations, the whole body DF is the most restrictive dose-based value, except for iodine which is governed by the dose to the thyroid (Tables 5 and 6). This is expected based on the known propensity of iodine to concentrate in the thyroid.

The influence of local climatology on MEI location and dose are illustrated in the heat maps in Figure 4. For the hypothetical Idaho facility, a main plume extends from the E toward the N with two smaller plumes extending S and SW. The hypothetical Tennessee facility has three nearly identical plumes, extending ENE, S, and SW. Clearly, the climatology and topography of these two locations are significantly different, such that transport and dose are unique to the region of the proposed facility.

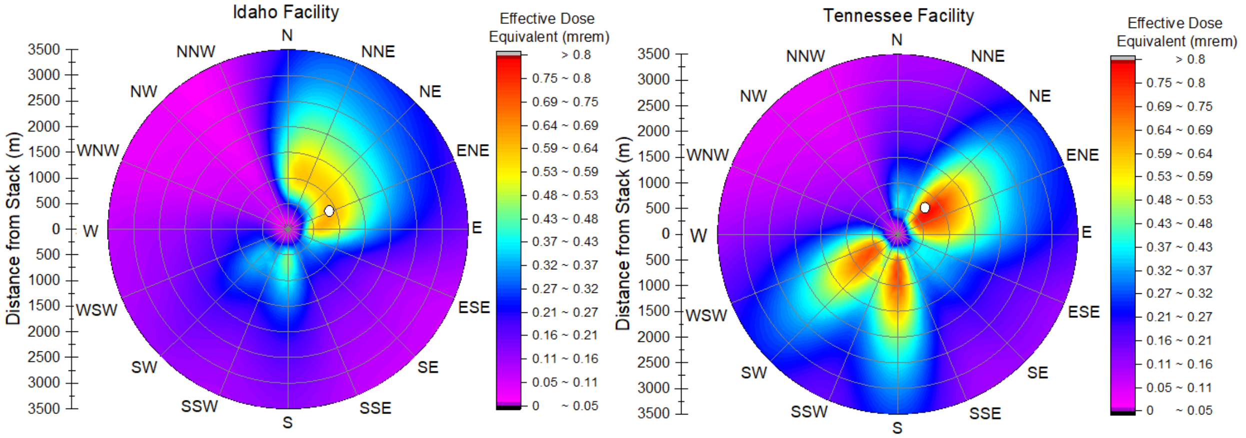

Figure 4: Comparison of effective dose equivalent as function of direction and distance from stack release in Idaho (left) and Tennessee (right) from base case of iodine vapor (200 MTIHM/y throughput, 50 m stack height, 12.2 m/s stack gas velocity, 2.2 m stack diameter). White circles represent location of predicted MEI. Dose mapping for each location is on the same scale.

The plots of radial location of dose shown in Figure 4 help explain the variability in direction and magnitude of the MEI observed in Tables 5 and 6. Heat maps for each isotope in this study for the base case are shown in Appendix A. For an individual location the plume shapes are identical for all isotopes, the only difference is the magnitude of the effective dose equivalents based on isotope. The location of MEIs in Tennessee at either due S or NE is likely due to minor differences in the magnitude of doses between the three plumes to the NE, S, and SW.

Overall, the climatological conditions at the two different locations have noteworthy effects on the dispersion of radionuclides from a stack and therefore, both locations are considered for all subsequent sensitivity analyses. The impact of climatological and topographical differences between the two locations is illustrated in Table 7, which shows the relative difference in DFs for each isotope between Tennessee and Idaho for a 200 MTIHM/y facility. These are based on the maximum DFs (whether whole-body or thyroid) as bolded in Tables 5 and 6. This comparison shows that the needed dose-based DFs can vary on the order of 20% for the chosen radionuclides based on different locations and meteorologies; except for tritium, which can vary on the order of 60%. For all isotopes, the dose-based DFs needed in Tennessee are higher than for the Idaho location because, at least in part, the MEI is closer to the facility than in Idaho. Other factors including precipitation and agricultural factors, may also have unique effects.

Table 7: Percent difference in effective dose for a facility located in Tennessee from the facility in Idaho for base case (200 MTIHM/y throughput, 50 m stack height, 12.2 m/s stack gas velocity, 2.2 m stack diameter). Maximum DFs are those identified in Tables 5 and 6 and result from either the whole body or thyroid dose.

Isotope	Maximum DF, Tennessee (10%)	Maximum DF, Idaho (10%)	Relative Difference (%)
Pu-238	1.8E+08	1.4E+08	22
Pu-239	3.5E+01	2.8E+01	22
Am-241	1.6E+03	1.3E+03	22
Cm-244	2.5E+08	2.0E+08	21
H-3	6.9E+00	1.1E+01	-60
Kr-85	5.0E-01	4.1E-01	18
I-129 (vapor)	2.0E+00	1.6E+00	19

4.1.3 Chemical and Physical Form of ^{129}I Sensitivity Study

The chemical and physical form of ^{129}I affects how it is incorporated into food cycles and can affect the dose from ^{129}I to the MEI, especially in the most conservative local agricultural case used our study. Iodine speciation can vary from more volatile species such as I_2, HI, HOI, and organic iodine species that may occur in aqueous used fuel reprocessing (Jubin et al. 2012) to less volatile iodides of Li, K, Na, and Cs that may be more likely in electrochemical reprocessing. CAP-88 allows for the choice of three input forms: particulate, vapor, or organic speciation. If particle phase speciation is selected both the size of the particle in microns and the absorption rate by the human body (fast, medium, or slow) can be specified. Tables 8 and 9 show that the choice of particulate, organic, or elemental vapor speciation has a large influence on the effective dose equivalent and required DFs. The particulate form delivers the largest dose (and therefore, largest DF values required) to an exposed individual, whereas the organic and vapor forms delivered much smaller doses. However, for both locations, there is no change in the effective dose to the MEI with particle size or the absorption rate of the particle (fast [F], medium [M], or slow[S]). As was observed in the base case scenario (Section 4.1.1) effective dose equivalents and corresponding DFs are higher for the hypothetical facility in Tennessee with the MEI located closer to the stack than Idaho (Tables 8 and 9). Heat maps of effective dose equivalents from iodine species as a function of direction and distance from the two hypothetical facilities are included in Appendix A.

Table 8: Summary of effective ^{129}I dose and location of MEI at a facility in Idaho and required DFs to meet EPA regulations. Particulate cases are modeled using fast [F], medium [M], or slow [S] slow adsorption rates.

Chemical Form	Location of MEI	Effective Dose Equivalent (mrem)	Thyroid Dose Equivalent (mrem)	Whole Body DF (10%)	Thyroid DF (10%)
0.1 micron (F)	600 m E	884	17300	3.5E+02	2.3E+03
0.1 micron (M)	600 m E	884	17300	3.5E+02	2.3E+03
0.1 micron (S)	600 m E	884	17300	3.5E+02	2.3E+03
1 micron (F)	600 m E	884	17300	3.5E+02	2.3E+03
1 micron (M)	600 m E	884	17300	3.5E+02	2.3E+03
1 micron (S)	600 m E	884	17300	3.5E+02	2.3E+03
10 micron (F)	600 m E	884	17300	3.5E+02	2.3E+03
10 micron (M)	600 m E	884	17300	3.5E+02	2.3E+03
10 micron (S)	600 m E	884	17300	3.5E+02	2.3E+03
Methyl Iodide	900 m ENE	0.67	9.32	2.7E-01	1.2E+00
Iodine Vapor	900 m ENE	0.60	11.90	2.4E-01	1.6E+00

Table 9: Summary of effective ^{129}I dose and location of MEI at a facility in Tennessee and required DFs to meet EPA regulations. Particulate cases are modeled using fast [F], medium [M], or slow [S] slow adsorption rates.

Chemical Form	Location of MEI	Effective Dose Equivalent (mrem)	Thyroid Dose Equivalent (mrem)	Whole Body DF (10%)	Thyroid DF (10%)
0.1 micron (F)	100 m S	1530	30000	6.1E+02	4.0E+03
0.1 micron (M)	100 m S	1530	30000	6.1E+02	4.0E+03
0.1 micron (S)	100 m S	1530	30000	6.1E+02	4.0E+03
1 micron (F)	100 m S	1530	30000	6.1E+02	4.0E+03
1 micron (M)	100 m S	1530	30000	6.1E+02	4.0E+03
1 micron (S)	100 m S	1530	30000	6.1E+02	4.0E+03
10 micron (F)	100 m S	1530	30000	6.1E+02	4.0E+03
10 micron (M)	100 m S	1530	30000	6.1E+02	4.0E+03
10 micron (S)	100 m S	1530	30000	6.1E+02	4.0E+03
Iodine Vapor	700 m NE	0.74	14.8	3.0E-01	2.0E+00

A sensitivity study on moving from 100% vapor to 100% particulate forms of iodine shows that the needed DFs scales linearly with the change in form (Figure 5). If most of the iodine is in particulate form such as in the form of iodides of Li, K, Na, and Cs, then needed iodine DFs (based on thyroid dose, which are higher than for the whole body) can become high as about 2,000. With increasing speciation of iodine to more volatile forms such as I_2 and ICl_x the needed iodine DFs could decrease to about 2. The iodine speciation can have a factor of up to a three order of magnitude difference on the needed iodine DFs. The speciation and physical form of any iodine that could be released from electrochemical reprocessing is not well understood and should be further evaluated to better assess what DFs are needed for iodine emissions. Given the effects of physical form on the required DFs, the distribution of iodine between particulate and vapor phase species is likely to be influential.

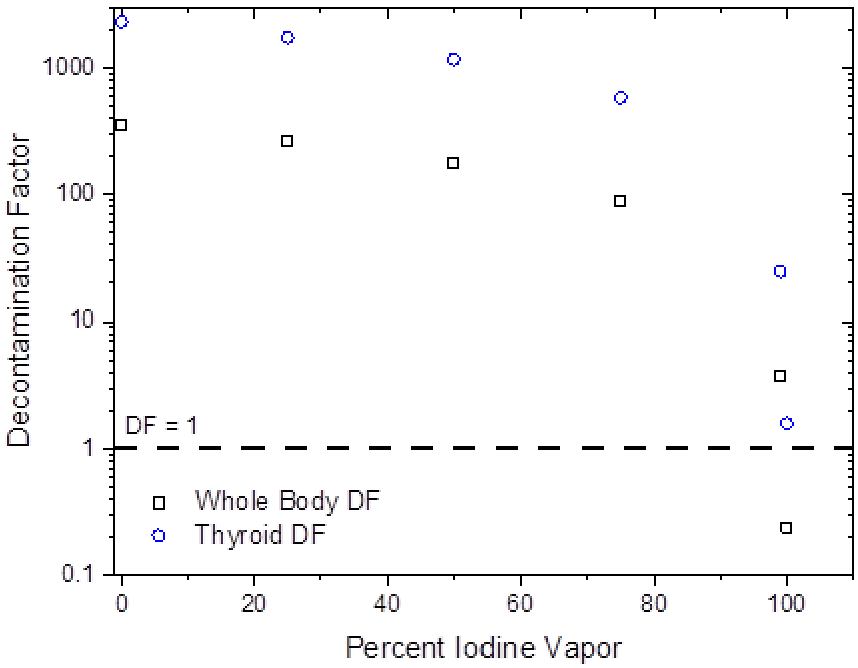

Figure 5: Semi-log plot of ^{129}I DFs as a function of iodine chemical form (vapor or particulate) for the hypothetical Idaho facility at base case conditions.

4.1.4 Stack Height Sensitivity Study

A stack height sensitivity study was performed for the hypothetical facilities in Idaho and Tennessee to identify how the height would affect the distance to the MEI and what consequences there would be to the DFs for ^{3}H, ^{85}Kr, ^{129}I, ^{239}Pu, and ^{241}Am. Stacks are normally used to elevate the emission of gasses from a facility to higher levels than surrounding buildings and to assist in the dispersion of the plume over a wider area. This, in theory, should translate into lower concentrations/doses delivered to the MEI and lower DFs.

Figures 6 and 7 and Tables A1 and A2 show the effect of changing the stack height for the hypothetical facilities in Idaho and Tennessee using the reference case used for the calculations done in this study. This sensitivity study did not incorporate meteorological data specific to each stack height; as such these figures are primarily illustrative. They identify that for both facilities as the stack height is increased, the dose to the MEI and required DF decrease. Increasing the stack height from 30 to 150 m (a possible stack height range for industrial processes) may change the needed DFs by 60–95% in Idaho and 63–97% in Tennessee, depending on the isotope. To completely analyze the effects of stack height on DF would require a significantly expanded study than the one presented here on both sites, which was beyond the scope of this study.

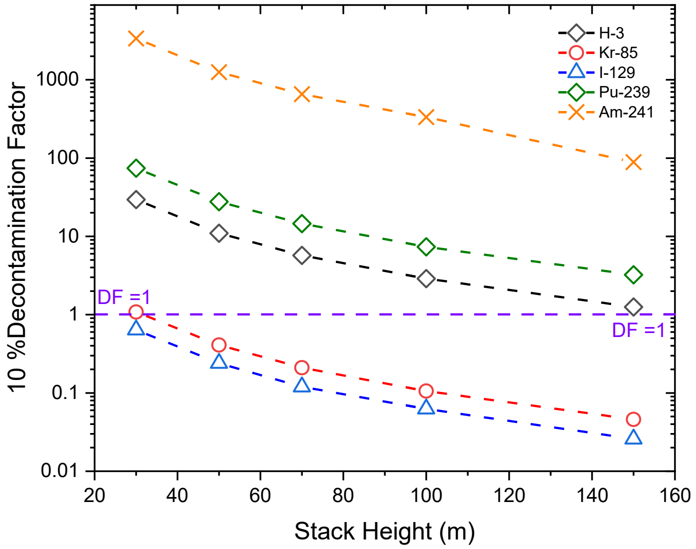

Figure 6: Stack height comparison for a facility in Idaho of DF as a function of stack height, for 200 ton/y throughput, 2.2 m stack diameter, 12.2 m/s stack velocity, and 2 y cooling time. Dotted lines are to represent trends only.

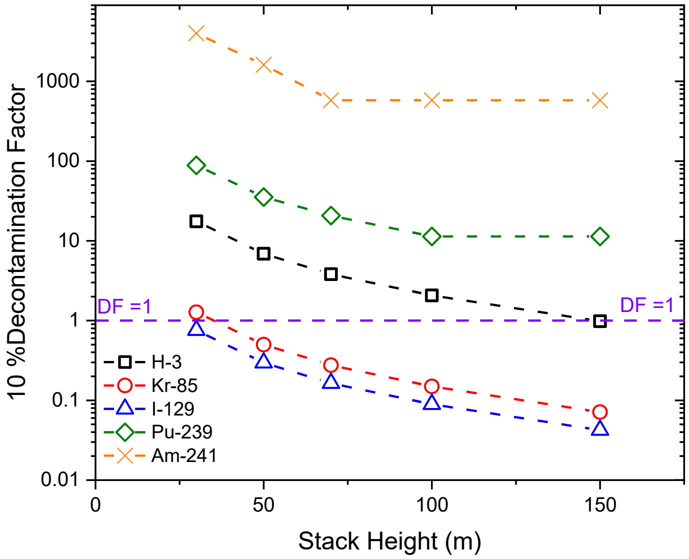

Figure 7: Stack height comparison for a facility in Tennessee of DF as a function of stack height, for 200 ton/y throughput, 2.2 m stack diameter, 12.2 m/s stack velocity, and 2 y cooling time. Dotted lines are to represent trends only.

Changing stack height alters DFs required for regulatory compliance and can also change the location of MEI and overall doses to the regions surrounding the stack. Figures 8 and 9 show the change in effective dose equivalent over the 3,500 m radius from the facility at stack heights ranging from 30 to 150 m in

Idaho (Figure 8) and Tennessee (Figure 9). Greater air dispersion at higher stack heights decreases doses across the entire region for both locations, not just for the MEI. For the Idaho facility, increasing stack height moves the location of the MEI from ENE to due E (Figure 8), but increasing stack height moves the MEI in Tennessee from NE to due S (Figure 9). Despite the change in MEI location, plume shapes are relatively maintained with increasing stack height.

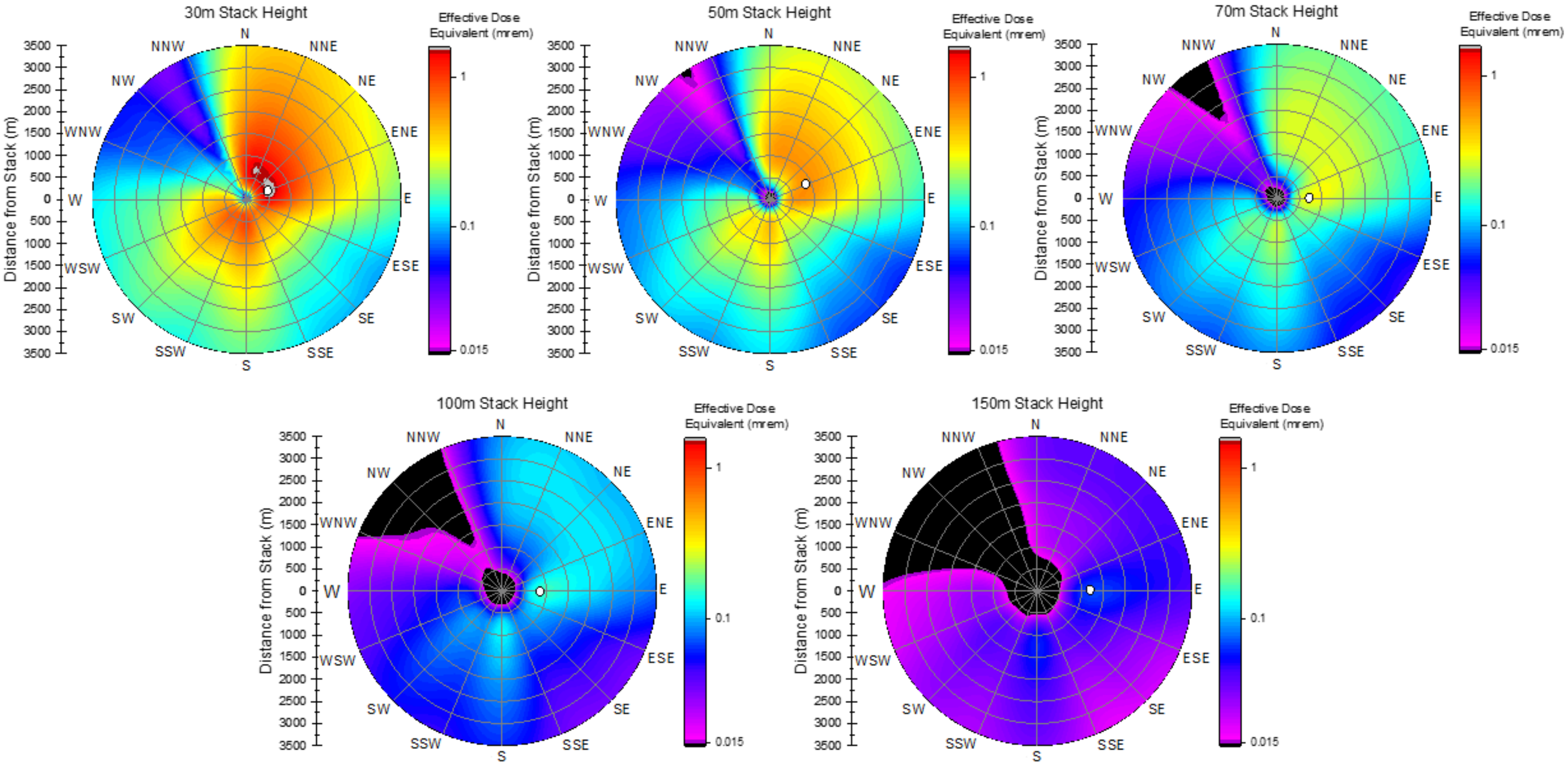

Figure 8: Heat maps meant to illustrate the potential changes in effective dose equivalent from iodine vapor as function of direction and distance from stack release in Idaho at varying stack heights. All other model parameters (stack gas velocity, stack diameter, throughput) are held constant at base case conditions. White circles represent location of predicted MEI, which is changing from ENE to E with increasing stack height and increasing in distance from 500 to 1,200 m from stack.

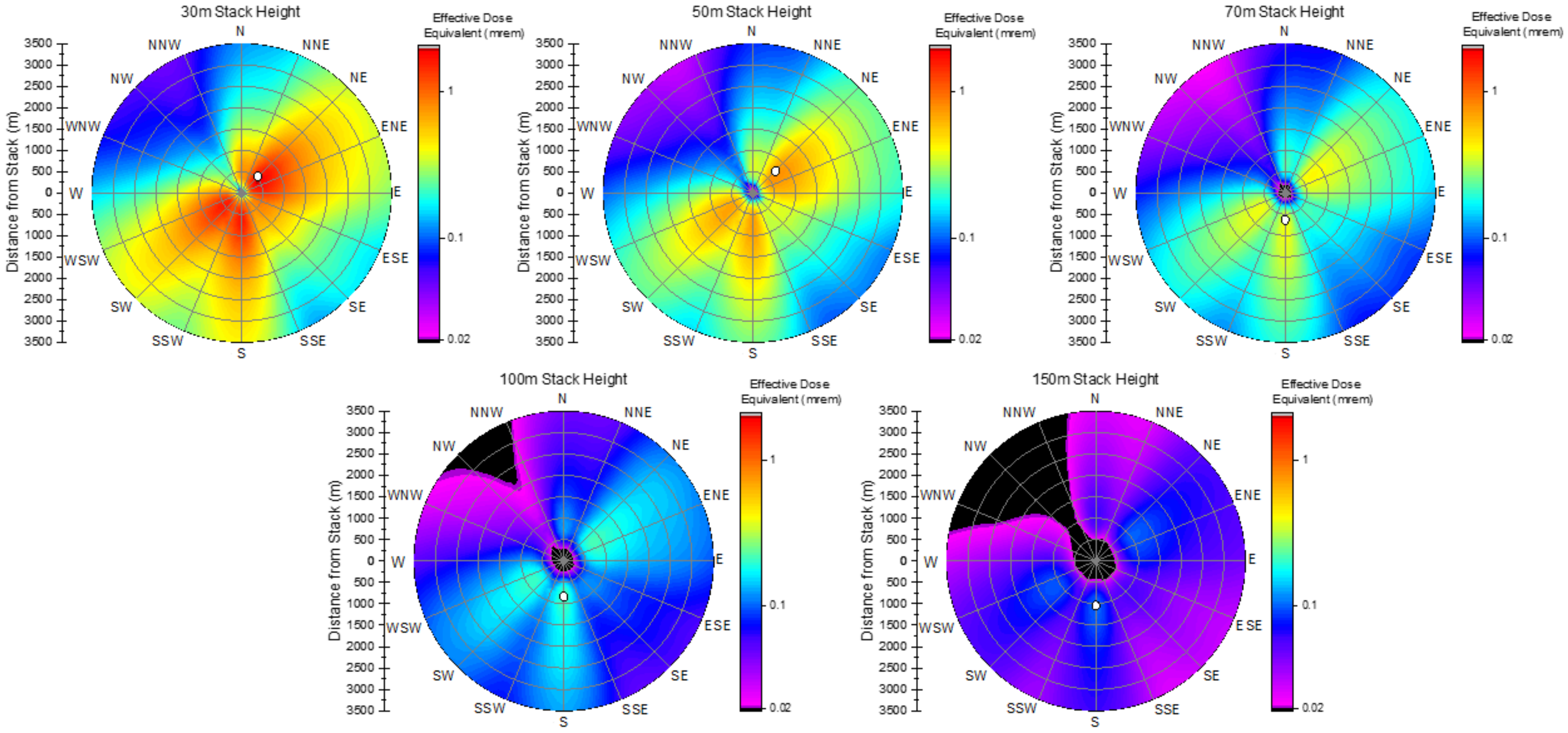

Figure 9: Heat maps meant to illustrate the potential changes in effective dose equivalent from iodine vapor as function of direction and distance from stack release in Tennessee at varying stack heights. All other model parameters (stack gas velocity, stack diameter, throughput) are held constant at base case conditions. White circles represent location of predicted MEI, which is changing from NE to S with increasing stack height and increasing in distance from 500 to 1,000 m from stack.

4.1.5 Stack Exit Gas Velocity Sensitivity Study

It is generally assumed that the higher the exit velocity of the stack gas, the higher the plume rises into the air. As with increasing the stack height, this should result in improved dispersion of the plume, increasing the distance to the MEI and lowering the dose. In this sensitivity analysis the stack height was kept at a constant 50 m, the stack velocity increased, but the stack diameter was decreased to maintain similar volumetric flow rates in the stack.

Tables 10–11 show the effect of increasing the stack gas exit velocity on dose and required DFs. Increasing the stack gas exit velocity from 6.2 to 18.2 m/s (a typical stack exit velocity range for industrial processes) can decrease needed DFs by about 20%–30% for Idaho and 9%–14% for the hypothetical facility in Tennessee, depending on the isotope. For example, the required ^{3}H DF for a 6.2 m/s stack exit gas velocity at the hypothetical Idaho facility is 12.2, and decreases to 10.6 for a stack gas velocity of 18.2 m/s. Thus, the variation of required DFs as a function of stack exit gas velocity is of significantly smaller magnitude than the variation of required DFs as a function of stack height.

Table 10: Impact of stack gas exit velocity on dose location and required DF for a facility located in Idaho.

Isotope	Stack gas velocity (m/s)	Stack diameter (m)	Location of MEI	Effective dose equivalent (mrem)	Whole body DF (10%)
H-3	6.2	2.7	600 m E	30.5	1.2E+01
Kr-85	6.2	2.7	600 m E	1.12	4.0E-01
I-129	6.2	2.7	600 m E	0.67	3.0E-01
Pu-239	6.2	2.7	500 m E	77.2	3.1E+01
Am-241	6.2	2.7	500 m E	3,490	1.4E+03
H-3	12.2	2.2	900 ENE	27.4	1.1E+01
Kr-85	12.2	2.2	900 ENE	1.02	4.0E-01
I-129	12.2	2.2	900 ENE	0.60	2.0E-01
Pu-239	12.2	2.2	900 ENE	69.3	2.8E+01
Am-241	12.2	2.2	900 ENE	3130	1.3E+03
H-3	18.2	1.7	900 ENE	26.4	1.1E+01
Kr-85	18.2	1.7	900 ENE	0.97	3.8E-01
I-129	18.2	1.7	900 ENE	0.58	2.3E-01
Pu-239	18.2	1.7	900 ENE	66.7	2.7E+01
Am-241	18.2	1.7	900 ENE	3,000	1.2E+03

Table 11: Impact of stack gas exit velocity on dose location and required DF for a facility located in Tennessee.

Isotope	Stack gas velocity (m/s)	Stack diameter (m)	Location of MEI	Effective dose equivalent (mrem)	DF (10%)
H-3	6.2	2.7	700 m S	20.8	8.3E+00
Kr-85	6.2	2.7	700 m S	1.5	6.0E-01
I-129	6.2	2.7	700 m S	0.849	3.4E-01
Pu-239	6.2	2.7	500 m S	104	4.2E+01
Am-241	6.2	2.7	400 m S	4840	1.9E+03
H-3	12.2	2.2	700 m NE	17.2	6.9E+00
Kr-85	12.2	2.2	700 m NE	1.25	5.0E-01
I-129	12.2	2.2	700 m NE	0.741	3.0E-01
Pu-239	12.2	2.2	500 m S	88.5	3.5E+01
Am-241	12.2	2.2	500 m S	4030	1.6E+03
H-3	18.2	1.7	700 m NE	16.3	6.5E+00
Kr-85	18.2	1.7	700 m NE	1.18	4.7E-01
I-129	18.2	1.7	700 m NE	0.704	2.8E-01
Pu-239	18.2	1.7	700 m NE	83.6	3.3E+01
Am-241	18.2	1.7	500 m S	3730	1.5E+03

4.1.6 Stack Exit Flow Rate Sensitivity Study

Like the stack gas velocity, it is generally assumed that the higher the exit flow rate of the stack gas, the higher the plume rises into the air. Further, because flow is a volumetric measure, increasing the overall flow decreases the concentrations of the radionuclides in the plume and should assist in dispersal of the dose from any radionuclides in the stack gas.

The results of the sensitivity study on the stack gas flow rate (Tables 12 and 13) show that increasing the stack gas flowrate has about the same effect on DFs as increasing the stack gas velocity. Depending on the isotope, increasing the stack gas flowrate from about 24 to 69 m^3/s may decrease the DFs by about 12%–64% in Idaho and 22%–67% in Tennessee.

Table 12: Comparison of dose, DF, and location of MEI as a function of stack gas flow rate for 200 MTIHM/y throughput, 50 m stack height for a facility located in Idaho.

Isotope	Stack gas velocity (m/s)	Stack diameter (m)	Volumetric flow rate (m^3/s)	Location of MEI	Effective dose equivalent (mrem)	DF (10%)
H-3	6.2	2.2	23.57	500 m E	31.80	1.3E+01
Kr-85	6.2	2.2	23.57	500 m E	1.16	4.6E-01
I-129	6.2	2.2	23.57	500 m E	0.69	2.8E-01
Pu-239	6.2	2.2	23.57	500 m E	80.30	3.2E+01
Am-241	6.2	2.2	23.57	500 m E	3620.00	1.4E+03
H-3	12.2	2.2	46.38	900 m ENE	27.40	1.1E+01
Kr-85	12.2	2.2	46.38	900 m ENE	1.02	4.1E-01
I-129	12.2	2.2	46.38	900 m ENE	0.60	2.4E-01
Pu-239	12.2	2.2	46.38	900 m ENE	69.30	2.8E+01
Am-241	12.2	2.2	46.38	900 m ENE	3130.00	1.3E+03
H-3	18.2	2.2	69.18	900 m ENE	24.30	9.7E+00
Kr-85	18.2	2.2	69.18	900 m ENE	0.89	3.6E-01
I-129	18.2	2.2	69.18	900 m ENE	0.53	2.1E-01
Pu-239	18.2	2.2	69.18	900 m ENE	28.80	1.2E+01
Am-241	18.2	2.2	69.18	900 m ENE	2770.00	1.2E+03

Table 13: Comparison of dose, DF, and location of MEI as a function of stack gas flow rate for 200 MTIHM/y throughput, 50 m stack height for a facility located in Tennessee.

Isotope	Stack gas velocity (m/s)	Stack diameter (m)	Volumetric flow rate (m^3/s)	Location of MEI	Effective dose equivalent (mrem)	DF (10%)
H-3	6.2	2.2	23.57	600 m S	22.1	8.9E+00
Kr-85	6.2	2.2	23.57	600 m S	1.6	6.4E-01
I-129	6.2	2.2	23.57	600 m S	0.952	3.8E-01
Pu-239	6.2	2.2	23.57	500 m S	114	4.6E+01
Am-241	6.2	2.2	23.57	400 m S	5190	2.1E+03
H-3	12.2	2.2	46.38	700 m NE	17.2	6.9E+00
Kr-85	12.2	2.2	46.38	700 m NE	1.25	5.0E-01
I-129	12.2	2.2	46.38	700 m NE	0.741	3.0E-01
Pu-239	12.2	2.2	46.38	500 m S	88.5	3.5E+01
Am-241	12.2	2.2	46.38	500 m S	4030	1.6E+03
H-3	18.2	2.2	69.18	800 m NE	14.7	5.9E+00
Kr-85	18.2	2.2	69.18	800 m NE	1.06	4.2E-01
I-129	18.2	2.2	69.18	800 m NE	0.632	2.5E-01
Pu-239	18.2	2.2	69.18	800 m NE	75	3.0E+01
Am-241	18.2	2.2	69.18	700 m NE	1700	6.8E+02

4.1.7 Initial Observations regarding Dose Rate and Required DFs

Several initial observations were made during the performance of the air dispersion modeling.

First, the climate and topography of the hypothetical facility location play an important role in the deposition of and dose from radionuclide emissions; therefore, two locations were considered throughout all dose estimations. In general, the MEI is at a further distance from the stack in the arid conditions of Idaho than for the more wet climate of Tennessee. Local topography and prevailing wind patterns likely explain the differences in plume shapes observed for the two locations. For both locations, dose mapping indicates that a single data point for a maximally exposed individual, although necessary for assessing regulatory compliance, does not fully capture the complexity involved with atmospheric transport and subsequent dose assessment of volatile radionuclides. Although consistent with regulatory language, the use of average annual meteorological inputs will reflect less precision in the annual dose to the MEI than a model that accounts for seasonal climate variability.

Second, the sensitivity studies presented here show that of potential facility parameters (stack height, stack gas velocity, stack gas flow rate), stack height has the most noticeable effect on required DF values for all isotopes. Increasing stack height is shown to increase plume dispersion and reduce dose to MEI (as well as doses across the entire region around the stack).

Third, the distribution of iodine between vapor and particulate forms will affect the calculated dose to the MEI (and the resultant iodine DF). Vapor phase species may be emitted from the facility, and these species will result in a relatively low dose to the MEI. However, if these vapor-phase iodine species undergo subsequent reactions with atmospheric gases or aerosols after their release they could form particulate phases. These particulate phases can result in relatively high doses to the MEI. The potential for this type of interaction should be considered. Within the parameters modeled, variation in particle size and adsorption rate did not have an impact on calculated DFs.

4.2 Emissions Limits Based on 40 CFR 190.10(b)

The facility DFs, shown in Table 14, required to meet the fuel cycle limits in 40 CFR 190.10(b) were calculated for the reference fuel case. These DFs depend only on the electricity produced by the fuel and the used fuel composition. The DFs were calculated using the following information:

- The burnup of the fuel is 99.6 $GW_{th}\cdot d/MTIHM$. The thermal efficiency for SFRs is expected to be roughly 40%, providing a conversion factor for GW_{th} to GW_e of 0.4 (IAEA 2012).
- The total activity of the relevant TRUs (including ^{239}Pu) is the sum of the TRU radionuclide activities shown in Table 3.
- The specific activity of ^{85}Kr is 391 Ci/g, and the specific activity of ^{129}I is 0.000176 Ci/g.

Table 14: DFs for ^{85}Kr, ^{129}I, and ^{239}Pu-TRU$_{<1y}$ as required by 40 CFR 190.10(b) for NRC-licensed commercial facilities.

Isotope	40 CFR 190.10(b) limit (Ci/GWe·y)	Activity of reference fuel (Ci/GWe·y)	DF
^{85}Kr	50,000	5.54E+04	2.8E+00
^{129}I	0.005	4.87E-01	2.4E+02
^{239}Pu-TRU$_{<1y}$	0.0005	1.21E+06	6.1E+09

For the selected case, the fuel cycle emission limits for ^{85}Kr, ^{129}I, and ^{239}Pu-TRU$_{<1y}$ result in DFs on the order of 3 for ^{85}Kr, 240 for ^{129}I, and 6.1E+9 for ^{239}Pu-TRU$_{<1y}$. As stated in Section 2, These limits only apply to NRC-licensed commercial facilities and are not relevant to DOE facilities such as the FCF at INL.

4.3 Throughput and Limiting Regulations

One of the key factors affecting the potential mitigation requirements for the emission of volatile radionuclides from an electrochemical reprocessing facility is that of facility throughput. As 40 CFR 190.10(b) limitations on ^{85}Kr, ^{129}I and ^{239}Pu-TRU$_{<1y}$ are based on the amount of electrical energy generated by the processed fuel, an increase in throughput rate would increase the releasable amount of each radionuclide, which results in a required fuel cycle-based DF that is constant regardless of the used fuel reprocessing rate. However, the dose-based restrictions in 40 CFR 190.10(a) are limits to an exposed individual. If all other factors (used fuel composition, stack height, meteorology etc.) are held constant, then the dose to the MEI and needed DFs vary proportionately with the used fuel reprocessing rate. This concept is illustrated by Figures 10 and 11.

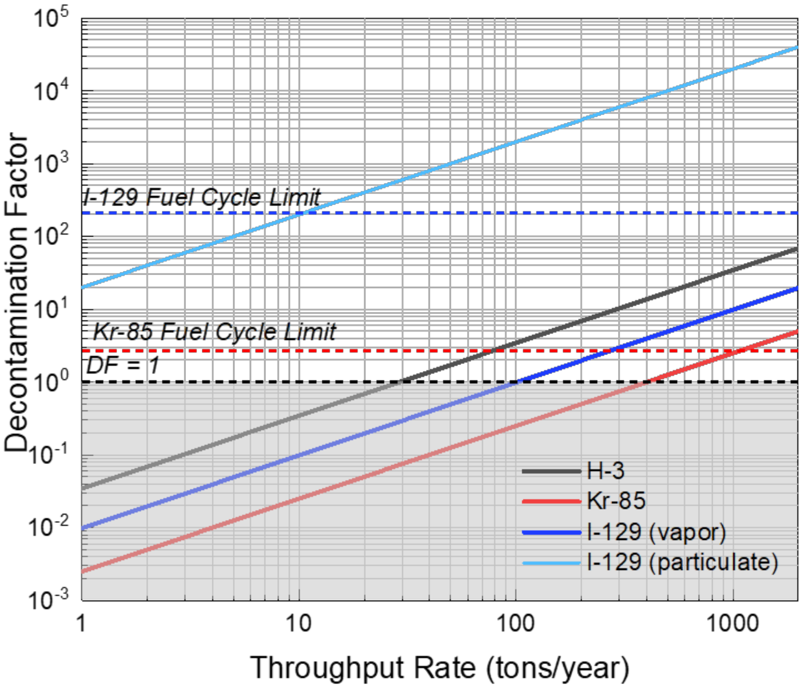

Figure 10: DF as a function of facility throughput rate for volatile radionuclides (^{3}H, ^{85}Kr, ^{129}I) for the base case at the Tennessee facility. Solid lines represent DFs based on 10% of allowable dose for the reference case and dashed lines represent DFs based on fuel cycle emission limits. Iodine DFs are for the limiting case of the thyroid dose.

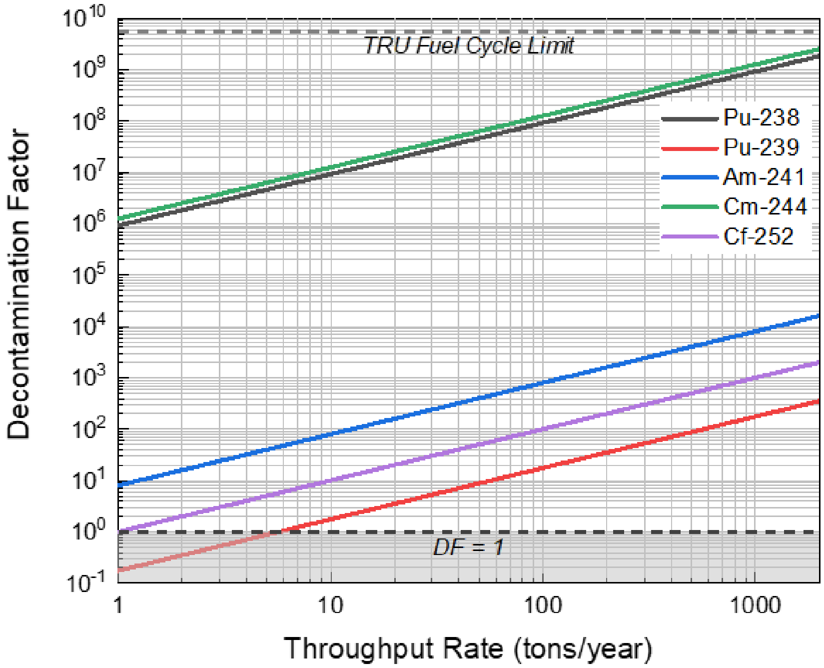

Figure 11: DF as a function of facility throughput rate for TRU isotopes for the base case at the Tennessee facility. Solid lines represent DFs based on 10% of allowable dose for the reference case, and dashed lines represent DFs based on fuel cycle emission limits.

These figures show that the emissions of each radionuclide can be governed by either 40 CFR 190.10(a) or 40 CFR 190.10(b). Tritium, which is not explicitly referenced in 40 CFR 190.10(b), is exclusively governed by dose-based limits, and the required DF for the reference case (assuming a 10% dose allocation) ranges from <1 below a throughput of 30 MTIHM/y to 7 at a throughput of 200 MTIHM/y. Similarly, in this reference case ^{85}Kr is governed by the fuel cycle limits of 40 CFR 190.10(b) unless throughput rates approach 1,000 MTIHM/y. The TRU radionuclides examined are controlled by the fuel cycle limits for all throughputs examined.

The governing regulations for iodine release under the reference case are variable based on throughput and physical form. Particulate iodine becomes governed by dose-based limits at throughputs above 10 MTIHM/y. Vapor-phase iodine species become governed by dose-based limits at throughputs above 100 MTIHM/y.

Section 6 examines the potential for the unit operations included in electrochemical reprocessing to provide the required mitigation for the relevant radionuclides and assesses whether additional off-gas controls are merited.

5. ELECTROCHEMICAL FUEL SEPARATIONS SYSTEM

Electrochemical reprocessing of used metallic fuel from sodium fast reactors includes chopping fuel elements into segments to expose the fuel meat to dissolution in molten salt, and several high-temperature processes used to separate fuel meat from the cladding segments, separate fission products from recyclable uranium–zirconium (U/Zr) and uranium–transuranic (U/TRU) products, and process the product and waste streams. Two primary waste streams are produced: a metal waste form (MWF) containing the used anode basket, remaining cladding, plenums, and undissolved Zr and noble metal fission products and a ceramic waste form (CWF) containing actinide-depleted, fission product-concentrated waste salt stream processed for disposal in a glass-bonded sodalite ceramic waste form. Chopping, separations, and waste form production are all performed in inert gas (Ar) hot cells designed to eliminate potential pyrophoric reactions of the used fuel, contain radioactive contamination, and shield workers and the environment from radiation exposure. Designs for electrochemical reprocessing facilities have included multiple unit operations within a single argon-covered hot cell, which can result in releases from multiple unit operations into a single large volume of inert gas.

Fuel element chopping is expected to be done at hot cell ambient temperatures on the order of 30°C. The various furnace temperatures can range from 500 to 1,700°C. Some radionuclides may evolve to the argon cell gas depending on speciation, retention in the chopped fuel, volatility of those species at cell and furnace temperatures, and solubility in molten LiCl–KCl eutectic salt.

Figure 12 shows the primary subsystems of this example electrochemical fuel reprocessing system. This electrochemical reprocessing system operates semi-continuously, with batches of used fuel entering the system and batches of separated product and waste material exiting the system. The product and waste materials will generally be composites from several batches of used fuel.

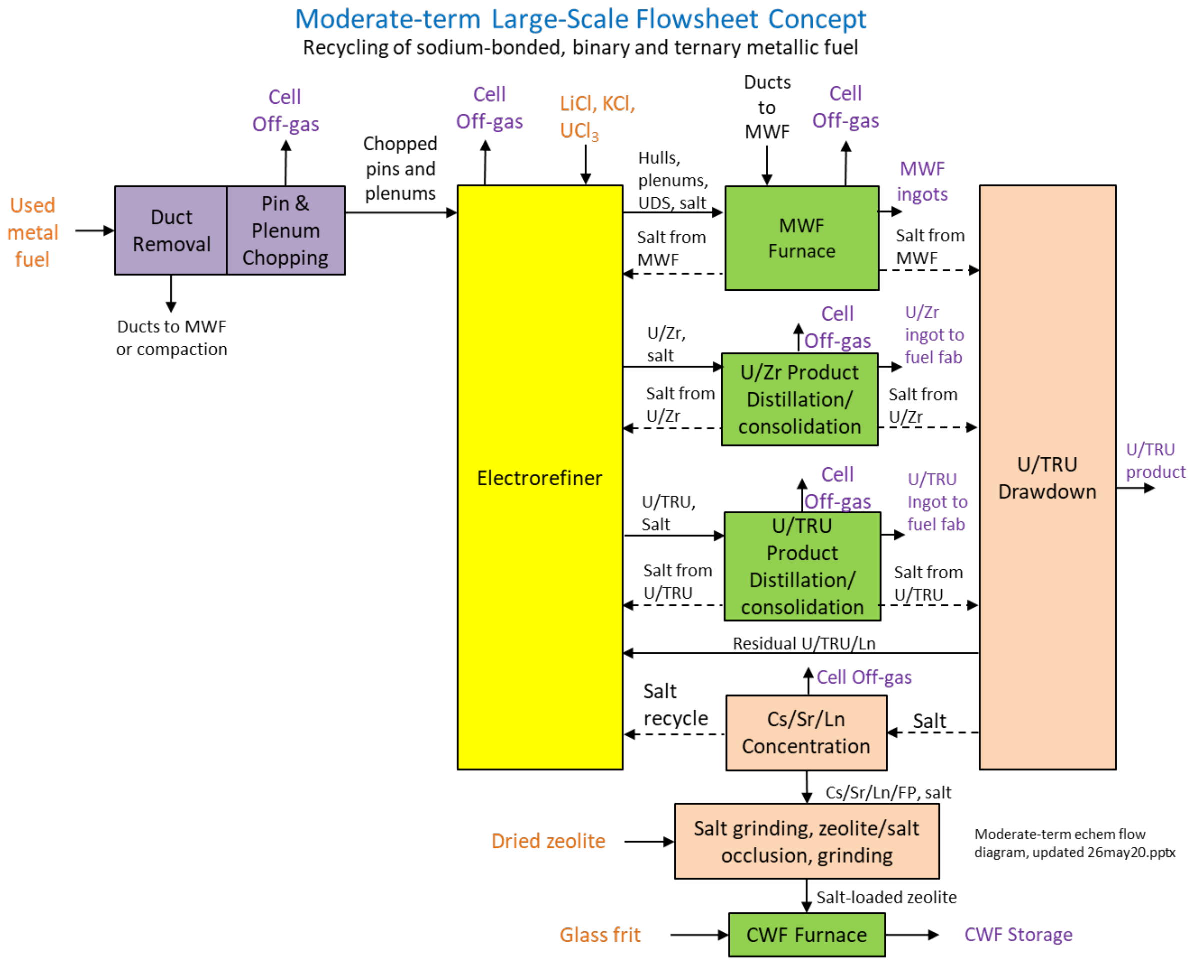

Figure 12: Electrochemical separations block flow diagram (adapted from Marsden 2020 and Dixon et al. 2019).

5.1 Head-End Operations

The head-end operations include receipt of used metallic fuel assemblies. The stainless-steel duct and other hardware that does not contact the actual fuel slugs are removed and can be separately compacted for disposal or melted into MWF ingots with the stainless-steel cladding and other metallic undissolved solids after the fuel is dissolved in the ER. These non-fuel-bearing components are not expected to have significant radioactive contamination (although they can contain activation products), so they would not likely be routinely processed in the ER.

The fuel pins with the plenums are mechanically chopped into segments on the order of 1.3–2.5 cm long. Any elements that tend to be volatile at ambient hot cell chopping temperatures of nominally 30°C (or higher depending on decay heat of the fuel itself) could be released to the cell gas. The extent that volatile elements are released to the cell gas depends on their speciation, migration into the plenums, and diffusivity of the chopped fuel segments.

5.2 Electrorefiner Subsystem

As generally described by Goff et al. (2011), Law et al. (2015), and Dixon et al. (2019), the chopped pins and plenums from the fuel chopping subsystem are placed in stainless-steel baskets and immersed in molten salt in the ER. The chopped fuel segments in the fuel basket function as the anode of the electrochemical cell, and an inert rod or solvent metal serves as the cathode of the cell. The process is conducted at 500–550°C in a LiCl–KCl eutectic melt that contains approximately 3–8 wt% dissolved uranium as uranium trichloride. As an electrical potential is applied to the cell, uranium contained in the fuel oxidizes and anodically dissolves to form soluble trivalent uranium ions that dissolve into the molten salt electrolyte. TRU elements; bond sodium; and lanthanide, alkali, and alkaline earth fission products also oxidize and anodically dissolve into the salt. Noble metal fission products such as Tc, Mo, and Ru, stainless-steel cladding and plenums, and the stainless-steel fuel basket do not dissolve under normal ER conditions because they are more noble (more electronegative) than uranium. These noble elements form chlorides with lower free energies of formation (and so are less stable) than uranium trichloride and are not oxidized at the operating potential of the cell. The electronegativity of zirconium is close enough to uranium so that some of the zirconium in the fuel meat dissolves under normal ER conditions.

Iodine and other elements that commonly form anions such as selenium and tellurium tend to dissolve into the molten salt during the fuel dissolution process and remain as anions in the salt throughout the refining process. Volatile elements including Kr, Xe, tritium (if present as a volatile species), and possibly iodine (if present as a volatile species) that did not already volatilize after chopping would be expected to volatilize into the cell gas as the fuel meat is heated and dissolved into the molten salt at 500°C.

As the fuel meat at the anode oxidizes and dissolves into the molten salt, the uranium, other actinides, and zirconium that dissolves from the fuel meat can migrate through the molten salt to the cathodes, where they can be electrolytically reduced and selectively deposited onto cathodes by controlling the electrochemical cell and cathode conditions. This process enables the separation of uranium in a solid U/Zr dendrite product collected on an inert cathode. A U/TRU product can be collected using a solvent metal cathode such as cadmium, which is molten at the ER operating temperature, that alloys with uranium and TRU elements (with typically small impurities from lanthanides).

5.3 U/Zr Product Purification Subsystem

The electrorefining process can separate U/Zr and U/TRU products from the used fuel and leaves the waste fission products dissolved in the molten salt. These product masses, however, can tend to have up to 20 wt% entrained salt impurity that adheres upon removal from the ER (and, in the case of the U/TRU product, the cadmium). The impure U/Zr and U/TRU products are each passed from the ER to separate U/Zr and U/TRU distillation and consolidation furnaces.

The U/Zr product is loaded into a zirconia-coated graphite crucible or zirconia crucible lining a graphite crucible before the treatment process. In the first step of the process, the bulk of the salt is vaporized from the uranium dendrites in the temperature range of 1,000–1,100°C and under reduced pressure (<1 torr). The salt vapor is cooled and condensed in the receiver of this furnace and ultimately transferred back to the ER or to a U/TRU drawdown furnace after a complete treatment cycle. During the second stage of the process, the temperature of the system is raised to approximately 1,200–1,400°C so that the U/Zr metal melts and any remaining salt occluded in the dendrites is vaporized and then cooled and condensed in the receiver. After the second heating period, the uranium is allowed to solidify into an ingot and cool to room temperature, removed from the crucible, and transferred to storage or reused in fuel fabrication. The crucible can be recoated and reused, or if it is damaged, it can be discarded as waste.

Similar to the salt removal from U/Zr product, cadmium and trace salt contamination is volatilized from the U/TRU product in a separate furnace. The crucible containing the cadmium cathode is loaded into a distillation furnace, and the temperature of the system increased to approximately 500–600°C under reduced pressure, which causes the cadmium to vaporize from the U/TRU product. The cadmium vapor is cooled and condensed in the receiver of the distillation furnace and is recycled to the ER after the process is complete. Salt contamination can be vaporized and separately condensed during this distillation process and returned to the ER or transferred to the U/TRU drawdown furnace. The U/TRU product is further heated until molten and free of cadmium and salt contaminants at temperatures up to 1,400°C, and then it is cooled and solidified into an ingot for transfer to storage or fuel fabrication. The crucible can be reused, or if it is damaged it can be discarded as waste.

5.4 Metal Waste Form Furnace Subsystem

Upon completion of an anodic dissolution cycle in the ER, the anode basket with remaining cladding, plenums, and undissolved Zr and noble metal fission products is removed from the ER and drained of molten salt. These materials, still containing approximately 20 wt% residual salt, are removed from the fuel baskets and transferred to a ceramic lined (e.g., alumina with yttria coating) graphite crucible. If additional iron or chromium is needed to produce the desired composition of the metal waste form (nominally 85 wt% stainless steel and 15 wt% Zr), then some stainless-steel duct from the used fuel assemblies or other material is added to the crucible. The crucible is placed in the MWF furnace, which is operated similarly to the U/Zr product furnace. Under reduced pressure, the temperature of the furnace is raised to approximately 1,000–1,100°C to allow the residual salt to vaporize from the noble metals and other materials. After the salt has been volatilized from the metals, the temperature of the system is increased to approximately 1,700°C to melt the metals, and then the metal is cooled and solidified into an ingot. The ingot is removed from the crucible and managed as high-level waste. Salt condensed and collected in the receiver of the distillation unit is transferred to the ER or to the U/TRU drawdown furnace. A small fraction of residual fission gases may be released to the cell gas during this process, if not already released earlier.

5.5 Salt Recycle Subsystem

The salt recycle subsystem in this example electrochemical process includes U/TRU drawdown and Cs/Sr/Ln concentration processes. As described in Law et al. (2015) the ER salt must be periodically purified to control the buildup of dissolved fission products that would otherwise cause excess impurities in the product streams. In the IFR concept and in the baseline INL electrochemical process, the salt was to be simply discarded and replaced with fresh salt (Priebe 2007, Priebe and Bateman 2008, Simpson et al. 2008, Morrison and Bateman 2010, Morrison et al. 2010). Several processes were considered for recovering the reuseable actinides dissolved in the spent salt before discarding it. This example electrochemical process uses electrolysis to reduce the actinides (dissolved in the molten salt) at a cathode to metal (which is deposited on the cathode). Chlorine gas is evolved at the anode (LaPlace et al. 2008 and Willit and Williamson, 2012). The actinide metal is recovered, and the actinide-depleted salt with fission products is processed further for recycling or disposal.

Even with the actinides separated and recovered from the recycle salt, the amount of salt that would need to be discarded because it contains some dissolved fission products would result in a relatively large volume and mass when converted to a durable salt waste form (Law et al. 2015, Vienna 2015a). Several process cases were evaluated in Vienna 2015b to reduce the eventual amount of the salt waste form. Technologies that have been studied to separate the fission product-laden salt into higher and lower fission product concentration salt streams include (a) electrolysis to electrolytically reduce LN chlorides to LN metal, (b) Precipitation of LN oxides by oxygen sparging (Frank 2011a, Choi et al. 2014), (c) Precipitation of LN as phosphates (Volkovich, et al. 2003), (d) ion exchange active fission product in zeolite (Ackerman, et al. 1997, Pereira, et al. 1999, Simpson et al. 2007, and Simpson 2013) (e) ion-selective membrane for separating Cs, Sr, Rb, and Ba (Spoerke et al. 2013, Spoerke et al. 2014), and (f) selective crystallization for separating Cs, Sr, and other fission products (Simpson 2013).

Based on these potential technologies, the example electrochemical process includes the concentration of fission products Cs, Sr, and lanthanides into a salt waste stream for further treatment disposal, and a salt recycle stream with depleted levels of fission products that can be recycled to the ER.

5.6 Waste Salt Glass-bonded Sodalite Ceramic Waste Form Subsystem

The actinide-depleted, fission product-concentrated waste salt stream in this example is processed for disposal in a glass-bonded sodalite ceramic waste form (CWF, Vienna 2015b). This process includes:

- Crushing and then grinding the waste salt at the ambient hot cell temperature to a particle size less than 250 μm.
- Blending the ground salt with dried zeolite while heating to ~500°C for ~80 h to occlude the salt onto the zeolite.
- Cooling, grinding, and blending the salt-loaded zeolite (SLZ) with a low-temperature glass frit at ambient hot cell temperature.
- Heating the SLZ-glass frit mixture in its waste canister to 925°C in stages and for sufficient time durations (~60 h at 500°C to equilibrate the temperature before the glass softens, and then ~75 h at 925°C) for all the components to react and form the desired glass-bonded sodalite waste form.

The final step is a controlled cool down intended to avoid cracking of the CWF block during cooling.

Alternative waste forms for the fission products have also been studied. These alternatives, including silica-alumina-phosphate (SAP) composite and Fe–P–O (iron phosphate) glass waste forms, have been studied to reduce the amount, or improve the durability, of the fission product waste form, and enable reuse of the chlorine in the salt (Frank et al. 2015, Riley et al. 2019).

5.7 Cell Gas Handling System

As discussed previously, it is expected that multiple unit operations will be conducted in a single hot cell and that these operations may vent directly into the hot cell. This design choice will result in the loss of discrimination between the emissions from individual unit operations. The hot cell is expected to use Ar as the cell gas and this gas will be routinely cleaned and recycled.

Species may evolve as gases at elevated furnace temperatures and vacuums (that can range from 500 to 1,700°C at vacuums up to 1 torr) and then cool to nominal cell gas temperatures of around 30°C, at nominal cell gas pressures of just under 1 atm. In order to control contamination within the hot cell, hot cell pressures are maintained at a slight (inches water) vacuum relative to the operating corridors, which are operated at a slight vacuum (inches water) relative to the ambient pressure.

As radionuclides that are gaseous at elevated furnace temperatures and furnace vacuums (such as radionuclides dissolved in ER salts) cool in the condenser regions of the distillation furnaces, they condense along with the ER salt. These condensed radionuclides are passed with the condensed salts either back to the ER or to the U/TRU drawdown furnace. The majority of radionuclides that are not captured in distillation furnace receivers eventually condense and form particulate matter if released into the nominally ambient temperature and pressure cell gas.

Depending on speciation, isotopes of some elements will not condense upon entering the nominally ambient temperature and pressure cell gas. These include noble gases Kr and Xe, tritium if in diatomic or tritiated water forms, and iodine if in forms such as I_2. I_2 specifically may tend to evolve with Cl_2 at the cathodes of electrolytic processes such as the U/TRU and LN drawdown processes.

Over time, the uncondensed gases and particulate matter that contain radionuclides build up in the cell gas. The particulate matter can tend to settle and deposit inside the hot cell.

The hot cell is expected to be equipped with a cell atmosphere handling system designed to maintain the cell gas temperature and pressure within target ranges, and maintain levels of particulate matter (dust) and gas contaminants (including both gaseous radionuclides and also oxygen and moisture from air contamination) within nominal levels (Lineberry et al. 1985, Law et al. 2015, Vienna et al. 2015, Marsden and Frank 2020). Figure 13 illustrates some features that a cell atmosphere handling system may need, depending on control requirements. These include

1. high-efficiency particle air (HEPA) filtration,
2. catalytic conversion of inleaked O_2 and, if needed, $^3H_2$, to water,
3. water removal by molecular sieve,
4. noble gas capture if needed, and
5. additional HEPA filtration.

This conceptual cell gas cleanup system takes a slip-stream of cell gas through a closed circuit of operations to efficiently filter, cool (or heat) the gas as needed, remove oxygen and moisture from air ingress, and remove, if needed, noble gases and gaseous forms of other radionuclides. This figure is for illustration only; specific designs may sequence some of the unit operations differently, or use different filtration or fan schemes, or may include additional or different control technologies.

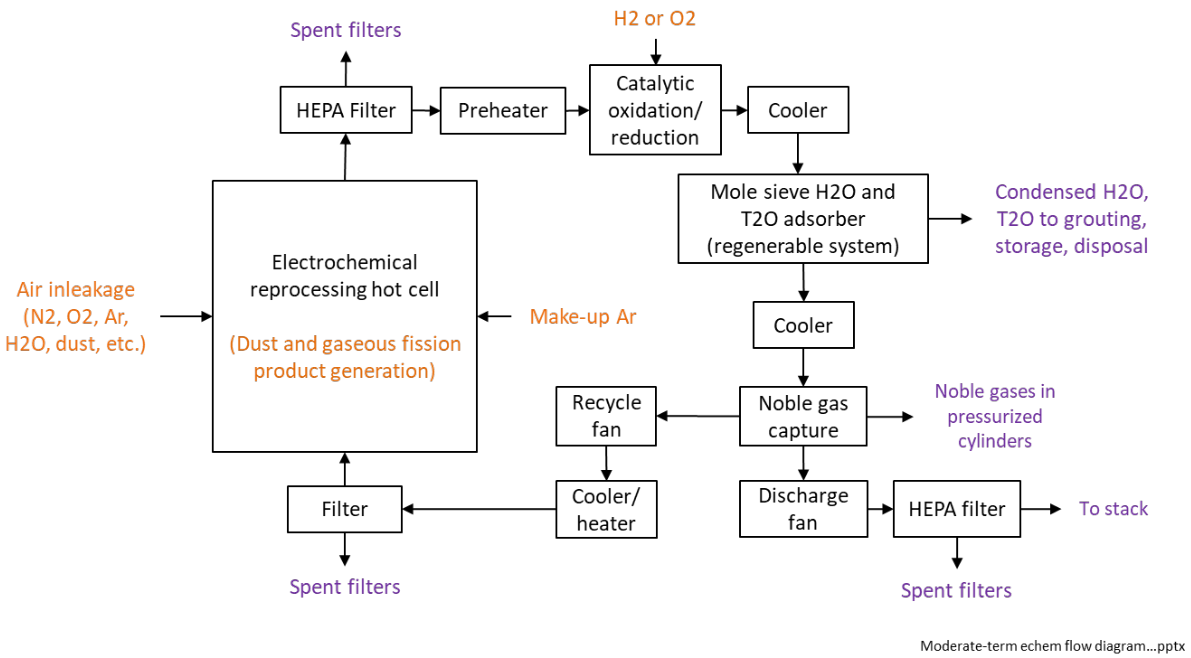

Figure 13: Illustration of an electrochemical reprocessing hot cell atmosphere handling system.

6. PARTITIONING OF REGULATED AND POTENTIALLY VOLATILE ISOTOPES DURING ELECTROCHEMICAL REPROCESSING

Figures 10 and 11 summarize the Section 4 estimates of overall facility DFs needed for a electrochemical reprocessing facility, based on the assumptions of used fuel composition, throughput rate, and other parameters that affect the dose to the public from radionuclides if they are emitted to the atmosphere.

That analysis is based upon complete release of the radionuclides from the fuel with no retention by the process and no intentional mitigation. Some of the radionuclides under consideration may have varying volatility, physical form, and chemical behavior that influence their potential retention within the facility. Some radionuclides can be nearly completely retained through standard facility design choices or through chemical interactions within the fuel processing operations, but other radionuclides (especially ^{3}H and ^{85}Kr) may not experience any retention and may quantitatively evolve to the facility off-gas. This section describes the expected behavior and fate of the volatile radionuclides within the facility and within the electrochemical reprocessing approach and how the needed DFs can be achieved for an electrochemical reprocessing facility.

6.1 Partitioning Assumed in the Case Study

Table 15 is adapted from the Case Study Report and summarizes the elemental partitioning that was assumed for electrochemical reprocessing by Law et al. (2015).

In the Case Study Report, 100% of ^{3}H (excluding the ^{3}H diffused into the coolant during SFR operation) and 100% of Kr were assumed to quantitatively volatilize into the cell gas during chopping, electrorefining, and MWF production. One hundred percent of the iodine was assumed to partition to the ER salt. One hundred percent of the ^{239}Pu-TRU$_{<1y}$ was assumed to partition primarily to the U/TRU product and secondarily to the MWF.

While these assumptions are sufficient for the general process description, they are not sufficient if even small amounts of atmospheric release could result in inability to comply with emissions regulations. For example, 40 CFR 190.10(b) could require a DF on the order of up to 200 for ^{129}I. This corresponds to a retention efficiency of 99.5% within the facility; release of >0.5% of ^{129}I from the facility stack would exceed regulatory limits. The ability to control iodine emissions with efficiency depends on iodine speciation in different parts of the process. Data with this level of precision was not found in a review of publicly available literature.

Table 15: Predicted elemental distribution within electrochemical reprocessing unit operations (adapted from Law et al. 2015). Green shaded boxes identify elements of concern for this report; orange shaded boxes indicate where they are released to the off-gas.

Subsystem	Element	Input Stream	Separation Factor	Discharge Stream
Head-end	Ducts, end pieces	Irradiated Fuel	1	Compacted metal waste form
	Bond Na		0.60	Plenums; sent to ER
	Cs		0.15	
	Iodine		0.10	
	H (including ^{3}H)[a]		1	Located in fuel matrix; sent to ER
	Kr, Xe (Group 18)		0.75	Released to head-end cell off-gas
	^{239}Pu-TRU$_{<1y}$		1	Located in fuel matrix; sent to ER
	All others		1	Located in fuel matrix; sent to ER
Electrorefining	U	Chopped pins, plenums	0.8799	U product
			0.12	U/TRU product
			0.0001	MWF
	TRU (including ^{239}Pu-TRU$_{<1y}$)		0.9999	U/TRU product
			0.0001	MWF
	Kr, Xe		0.80	ER off-gas
			0.20	MWF furnace off-gas
	H (including ^{3}H)		1	ER off-gas
	Iodine		1	Salt
	Group 1, Group 2, LN, Ac, C, Cd, Cl, Se, Pa, Ra, Se,Th		1	Salt
	Ag, Cr, Fe, Ir, Mo, Nb, Ni, Pb, Pd, Rh, Ru, Sb, Sn, Tc, Zr		1	MWF
	Te		0.80	Salt
			0.20	MWF
	Note: Mass contribution from spent baskets is not considered for simplicity.			
U, U/TRU, and MWF distillation/consolidation	Salt	Carried with metal	1	Recovered and returned to ER or sent to U/TRU drawdown
Actinide drawdown	Actinides not including Ac, Pa, and Th	Input salt	1	Actinide metal product
	All other salt constituents including Ac, Pa, and Th		1	Salt
LN drawdown	LN not including Eu and Sm	Input salt	1	LN metal product
	All other salt constituents including Eu and Sm		1	Salt

Table 15 (continued): Predicted elemental distribution within electrochemical reprocessing unit operations (adapted from Law et al. 2015). Green shaded boxes identify elements of concern for this report; orange shaded boxes indicate where they are released to the off-gas.

Subsystem	Element	Input Stream	Separation Factor	Discharge Stream
Cs/Sr recovery and [Li,K]Cl recovery	Cs, Sr, Ba, Rb, Eu, Sm	Input salt	Varies up to 0.95 to achieve mass balance closure for these elements in the different cases.	Waste salt
	LiCl-KCl		Enough LiCl-KCl so the conc. of total Cs, Sr, Ba, Rb, Sm, and Eu chlorides is 0.55.	Waste salt

[a]70% of ^{3}H generated during irradiation diffuses out of the fuel and into the coolant before processing of the assemblies.

6.2 Fate of Tritium

Historical results from the operation of EBR-II demonstrated that 70% of the tritium produced during fuel irradiation evolved from the fuel, passed through the stainless-steel cladding and into the SFR liquid sodium coolant (Ebersole et al. 1971). The ER process is assumed to be a separate facility from the reactor; so that any dose caused by any gaseous release of tritium from the reactor is not included in dose calculations for the reprocessing facility. The dose-based DF required for tritium in the reference scenarios evaluated was determined to be about 10; decreasing the amount of tritium in the received fuel by 70% decreases the maximum DF that could be needed for ^{3}H to about 3.

Tritium can be expected to volatilize as tritiated hydrogen gas ($^3H_2$) or other species (e.g., ^{3}HCl, ^{3}HI) having sufficiently high vapor pressures at process temperatures. The potential for the formation of less-volatile tritium-bearing species, such as metal hydrides, is not known. A literature search did not identify any empirical measurements that would inform as to the speciation of the tritium released from electrochemical processing operations. A list of potential species that could be formed in the ER is shown in Table 16, along with their boiling points. This list is not fully inclusive but reflects a best guess based on process knowledge and melt composition. Although boiling points are not directly reflective of partitioning between the salt melt and the cell atmosphere, they can give a relative indication of the tendency of a species to be retained in the salt. In this case, with the exception of any metal hydrides that form, notable potential hydrogen-bearing species have boiling points low enough that their complete release would be expected from the ER. Until potential ^{3}H speciation during electrochemical reprocessing is better understood, it may be conservatively assumed that all ^{3}H remaining in the used fuel following reactor discharge is released to the off-gas.

Table 16: Tritiated hydrogen-bearing species potentially present in the ER.

Species	Boiling point (°C)
H_2	−252
HCl	−85
HI	−35
Metal hydrides (LiH, transition metal hydrides, UH_x, ZrH_x etc.)	Regarded as nonvolatile

6.3 Fate of Krypton

While hydrogen, krypton, and iodine are normally identified and evaluated as “volatile” in aqueous reprocessing, only hydrogen (including ^3H) and krypton are thought to be predominantly “volatile” in electrochemical reprocessing. It is widely assumed that the bulk of the noble gases are released across two unit operations (chopping and electrorefining). It has been shown that up to 80% of krypton produced within the fuel matrix migrates to the plenum of the fuel rod during irradiation–this would be released upon chopping (FRWG 2018, Lee et al. 2001, Grabaskas et al. 2016). The electrorefining operation, which dissolves the fuel into a salt melt at 500°C, is assumed to release an additional 16% of Kr, with the balance released during metal waste form production (Law et al. 2015). Given the nature of the noble gases, it is unlikely that process chemistry or particulate filtration will result in any retention of Kr; it can be assumed that both krypton and xenon are completely released to the cell gas during electrochemical reprocessing.

6.4 Fate of Iodine

Iodine may form metal halides or mixed halide species during irradiation and electrochemical reprocessing, which are less-volatile than elemental iodine (I_2). The fate of iodine in used metal fuels and during used metal fuel electrochemical reprocessing has been studied in recent years. The potential exists for iodine to be released to the cell gas as either particulate or vapor-phase species, and little is known regarding the potential distribution of iodine between these two forms.

Iodine may partition in the fuel pins between the fuel region (either in the fuel itself or in bond sodium) and to the gas plenums, where it may exist in the form of I_2 or iodides of Na, Cs, or other cations (Frank 2011b, Frank 2014). Measurements have indicated that the 10–15% of the iodine inventory is present in the plenum (Grabaskas et al. 2016). The release fraction of iodine during chopping has not been measured, but it can be assumed that I_2 found in the plenums may be sufficiently volatile as to be partially released into the cell gas during this operation.

Frank et al. (2011) determined the distribution of iodine between the plenums, bond sodium, and metallic fuel matrix from a fuel pin irradiated in EBR-II. Experiments were also performed to evaluate the release potential for iodine from the ER, salt distillation, and ceramic waste form production operations. The experiments intended to evaluate iodine release from the ER were performed at operating temperature but did not have electrical current applied through the salt during the experiment.

No significant iodine release was identified during these experiments. However, it is important to note that the precision of these experiments may not be sufficient to demonstrate that the combined retention of iodine during chopping, electrorefining, salt distillation and waste form production is high enough to achieve overall iodine DFs on the order of 200 (as dictated by 40 CFR 190.10(b)). Table 17 shows potential iodine-bearing species that may be formed in used fuel or during electrochemical reprocessing , Based on their boiling or decomposition points, and expected solubilities in the ER salt, species that could be volatile if formed include I_2, ICl_x, and HI. Species that are not sufficiently volatile may exist as particulate, and these include KI, LiI, etc.

Table 17: Boiling points of selected iodine species.

Species	Boiling point (°C)	Associated with:
I_2	184.4	All
ICl_3	97 (decomposes)	All excluding chopping
CsI	1,280	Chopping, Electrorefining
NaI	1,304	Chopping, Electrorefining
KI	1,323	All excluding chopping
HI	−35.5	All excluding chopping
LiI	1,171	All excluding chopping
UI_x	Considered nonvolatile below <2,000°C	All excluding chopping
PuI_3		All excluding chopping

The actual partitioning and volatility of iodine depends upon mass transfer, kinetics, and thermodynamics. The iodine that partitions to the salt is expected to behave similarly to the much larger mass of dissolved Cl^- ions. A small fraction of the dissolved iodide is expected to be reduced to gaseous I_2 along with chloride that is reduced to gaseous Cl_2 during electrolytic U/TRU drawdown (Vienna et al. 2015a). If electrolysis is also used during salt recycle, a small amount of additional gaseous I_2 could be formed along with more Cl_2. The Cl_2 and I_2 gases are captured for recycle to make more ER salt, or for disposal. Any I_2 that is not captured would evolve to the cell gas.

6.5 Fate of ^{239}Pu-TRU$_{<1y}$

The ^{239}Pu-TRU$_{<1y}$ isotopes are expected to exist in nonvolatile forms. Any volatile emissions of these radionuclides to the cell gas from the high-temperature electrochemical processing operations would likely be followed by their immediate sublimation at cell gas ambient temperatures to form particulates or dust within the cell.

7. MITIGATION STRATEGIES

If the regulated and volatile isotopes addressed in this study (^{3}H, ^{85}Kr, ^{129}I and ^{239}Pu-TRU$_{<1y}$) are not adequately retained in the electrochemical processes to meet regulatory limits, then additional isotope-specific mitigation strategies may be indicated.

7.1 Tritium

Gaseous tritium-bearing species that might be expected to be released from electrochemical reprocessing the ER include H_2, HCl, HI, and ICl_x. Removal of tritiated H_2 could proceed via the conceptual cell gas treatment design provided in Section 5 within the catalytic oxygen removal operation. In this scenario, tritiated H_2 could react with O_2 to form H_2O that is subsequently absorbed by regenerable molecular sieve. An example of this catalytic conversion is shown in Equations 1 and 2. In this example, copper metal is used to remove oxygen from the gas stream, forming CuO. CuO is regenerated to Cu through the conversion of H_2 (or tritiated hydrogen) to water.

$$Cu(s) + O_2(g) \rightarrow CuO(s)\ (\Delta G \sim -58.6\text{ kcal/mol @ }100°C) \quad (1)$$

$$CuO + H_2 \rightarrow Cu + H_2O(g)\ (\Delta G \sim -24.5\text{ kcal/mol @ }100°C) \quad (2)$$

Upon regeneration of the molecular sieve, the tritiated water is condensed and collected for conversion into a suitable waste form such as grout for storage, transport, and disposal. To ensure capture of tritiated H_2 and to ensure low levels of H_2 in the cell gas well below flammability limits, this process is controlled by using substoichiometric amounts of added H_2 reactant (relative to the amount of O_2) from only small containers of H_2 reactant (a small H_2 reactant source term) so that there is an excess of O_2, while maintaining the excess O_2 within acceptable cell gas limits.

The selection of copper or other metal catalysts for use in the catalytic oxygen removal operation could also impact the mitigation of tritiated HCl or tritiated HI, as illustrated by Equations 3 and 4.

$$HI(g) + Cu(s) \rightarrow CuI(s) + \tfrac{1}{2}H_2(g)\ (\Delta G \sim -31.1\ \text{kcal/mol @ } 100^\circ\text{C}) \tag{3}$$
$$HCl(g) + Cu(s) \rightarrow CuCl(s) + \tfrac{1}{2}H_2(g)\ (\Delta G \sim -9.2\ \text{kcal/mol @ } 100^\circ\text{C}) \tag{4}$$

In this example, HI or HCl is converted to solid CuI or CuCl and releases H_2 gas, which could then interact to form water as shown in Equation 2. This illustration shows that the behavior of the potential tritium-bearing halide species within the catalytic bed should be verified after selection of a specific oxygen removal operation. If this occurs, or if the catalytic oxidation process can be optimized to make this occur, then this provides a pathway for control and disposal of ^3H that is released in the form of hydrogen halides. However, to the extent that this occurs, then it converts the oxidation catalyst into a consumable material that will require replacement and disposal. Another potential benefit from this chemisorption of halogens on the oxidizer catalyst is that this may also capture ^{129}I if in the forms of I_2, HI, or ICl_x.

The practicality of this method for capturing both ^3H and ^{129}I if they are present in gaseous halogen or halide forms depends on such factors as (a) how the catalytic oxidizer can be configured for periodic catalyst replacement, (b) the efficiency of this process for ^3H and ^{129}I capture, (c) catalyst consumption rates and costs considering potential co-sorption of chlorides which may be present in larger concentrations than the halides, and (d) conversion of the spent catalyst to a waste form suitable for storage, transport, and disposal. However, to the extent that this process could be made practical, it could be a way in a single process to capture gaseous forms of both ^3H and ^{129}I to meet needed DFs.

If tritiated halogens do not react with the oxidizer catalyst as described above, or if consumption of the oxidizer catalyst is not desired, then the potential for adsorbing tritiated halogens onto a regenerable molecular sieve (perhaps upstream of the catalytic oxidizer) could be evaluated. The potential exists to selectively remove tritiated HI and HCl via molecular sieves. Zeolite-based molecular sieves have great affinity for polar molecules (e.g., HCl) (Millipore Sigma 2020). Eng and Greenwood recommend a zeolite with a 5 Å aperture for sorbing HCl from a Cl_2 product. Berl (1961) provides a graphic for the HCl capacity of both 5 Å and 13X molecular sieves as shown in Figure 14. Capacity of about 13–21 wt% is indicated at a temperature of 25°C, but only at very high gas phase HCl concentrations. Additional data are needed on how these molecular sieves (or at least one of them) perform at lower HCl (and HI) concentrations. Cooling of the molecular sieve trap may improve its performance, but additional data is required to verify this approach.

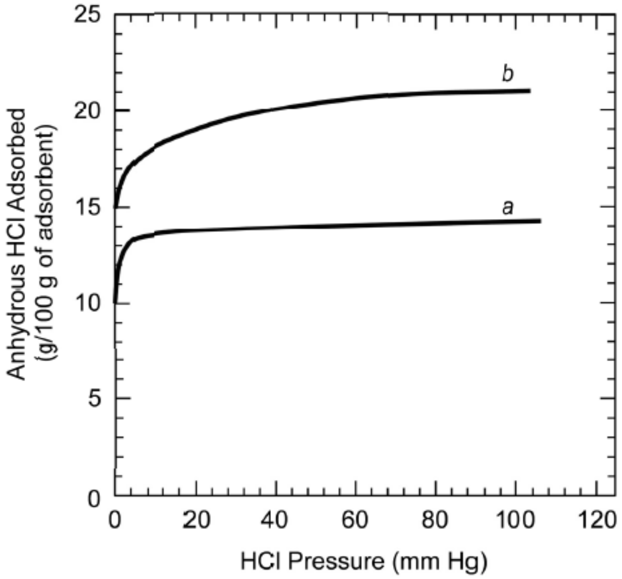

Figure 14: Adsorption capacity of molecular sieves (a) type 5 Å and (b) type 13X to trap HCl as a function of the partial pressure of HCl at 25°C (adapted from Berl 1961).

Other methods for capturing tritium in the form of hydrogen halides include activated carbon sorbents. The capacity of such sorbents needs to be evaluated to determine their practicality in this application.

7.2 ^{85}Kr

Two leading options for ^{85}Kr capture are cryogenic distillation of noble gases (such as described in Lineberry et al. 1985) and noble gas adsorption on solid sorbents at higher-than-cryogenic temperatures (Soelberg et al. 2015, Jubin et al. 2016, Bruffey et al. 2020). Adsorption of noble gases via solid sorbent is likely to require fewer gas pretreatment operations and be less energy intensive but is less well-developed than cryogenic separations.

Either technology can provide separation of the noble gases, which can be an important aspect of nuclear waste management. If the noble gases emitted from a hypothetical 200 MTIHM/y reprocessing facility are condensed and stored together in pressurized gas cylinders, roughly 240 Type 1A cylinders (internal volume of 43.8 L; pressurized to 5 MPa) would be produced per year. Removal of non-radioactive Xe, roughly 10× more abundant than Kr in irradiated fuel, would reduce this source of radioactive waste generation to 20 cylinders per year. DOE-NE is actively pursuing two types of solid sorbent technologies as alternatives to cryogenic distillation; the first is based on engineered silver- and hydrogen-reduced mordenite minerals (silver mordenite–polyacrylonitrile and hydrogen mordenite in a polyacrylonitrile-based engineered form) and the second is based upon the use of metal organic framework materials. The application of solid sorbents to noble gas separation from the cell gas of an electrochemical reprocessing facility is discussed in Section 9.

7.3 Iodine

The removal of iodine from gas streams depends on its physical form and/or chemical speciation. Particulate filtration is discussed in more detail in Section 7.4. Vapor-phase iodine removal is discussed here.

If vapor-phase iodine mitigation is needed, a few iodine control options may be considered. First, as introduced in Section 7.1, halogen species may react with the catalyst used in oxygen removal. Additional potential interactions are shown by Equations 5 and 6. Similar reactions could be envisioned for other common catalysts.

$$2Cu + Cl_2 \rightarrow 2CuCl(s)\ (\Delta G \sim -55.1\ \text{kcal/mole @ } 100°C) \tag{5}$$

$$2Cu + I_2 \rightarrow 2CuI(s)\ (\Delta G \sim -35.3\ \text{kcal/mole @ } 100°C) \tag{6}$$

This potential control strategy results in chemisorbed iodine (and chlorine if also present in the gas stream) that can be converted to a waste form suitable for storage, transport, and disposal. The catalyst is consumed.

Another potential iodine control option, for iodine in the form of I_2 or ICl_x, may be to cool the gas to a temperature low enough to condense or sublimate I_2 and ICl_x, as they have relatively high boiling points (Table 9). Evaluations would need to be done to determine, at the expectedly low iodine species concentrations, what low temperature could achieve the needed DFs.

A third option would be to use silver-based sorbents such as silver zeolite, silver-functionalized silica aerogel, or $AgNO_3$ on an alumina substrate to chemisorb the gaseous iodine (and chlorine) species. These and other silver-based sorbents have been evaluated by the Off-gas Sigma Team of the DOE Material Recovery and Waste Form Development Campaign for capturing inorganic and organic iodides from aqueous reprocessing off-gas streams (Soelberg et al. 2013, Jubin et al. 2016, and Bruffey et al. 2020). The use of oxidizer catalyst or silver-based sorbents for capturing iodine may not be desirous due to the chlorine likely to be associated with any vapor-phase iodine. Chlorine levels higher than iodine levels in the cell gas or process-specific off-gas could lead to higher consumption of silver-based materials due to the formation of AgCl. Given that silver is both a precious and hazardous metal, the consumption and waste generation arising from chlorine consumption of the sorbent may be undesirable. Alternative sorbent materials have been considered. These include activated charcoal, zeolites exchanged with alternative metals (Pb, Bi, Na), MgO particles, metal organic frameworks, or several other types of sorbents (Jubin 1979, Nandanwar et al. 2016, Huve et al. 2018). However, absorption of iodine by silver-free sorbent materials has yet to be demonstrated as viable and economical for removal of iodine from the off-gas streams associated with nuclear fuel reprocessing.

A fourth option would be to incorporate iodine capture with solid sorbent noble gas capture. If cryogenic noble gas capture is used, then iodine-bearing species would need to be captured before the cryogenic noble gas process. If solid sorbents are used for noble gas capture, iodine species may interact with the solid sorbents selected for use. For example, in the case of AgZ–PAN, iodine and chlorine species may chemisorb to form AgI and AgCl. The structural changes that may occur as a result of this chemisorption could impact the noble gas separation itself.

7.4 ^{239}Pu-TRU$_{<1y}$

As discussed in Section 6.5, ^{239}Pu-TRU$_{<1y}$ isotopes are expected to exist as particulates if entrained into gas streams. The cell gas treatment system will include filtration, and this filtration can be used to comply with the established regulatory emissions limits.

The control efficiency of particulate-form radionuclides can be conservatively estimated using the US EPA's strategy in 40 CFR 61 Appendix D – Methods for Estimating Radionuclide Emissions. This strategy conservatively applies emission factors (which are the inverse of DFs) of 1 for gaseous radionuclides; 10^{-3} (0.1% or 1,000 DF) for liquid or particulate solids; and 10^{-6} (0.0001% or 10^6 DF) for solids. If any nuclide is heated to 100°C or more, boils at 100°C or less, or is intentionally dispersed into the environment, it must be considered to be a gas. For particulates, the strategy allows an adjustment factor of 0.1 (10 wt% or 10 DF) for each fabric filter, and 0.01 (1% or 100 DF) for each HEPA filter, used in the off-gas system.

All of the elements in the used fuel that do not volatilize before entering the ER are heated to over 100°C in the ER, and so are conservatively assumed to have an attenuation of 1; but then cool to nominally ambient temperature in the cell gas, and so can be assumed to condense in the cell gas and exist in particulate form. An attenuation factor of 10^{-3} (0.1 wt% or 1,000 DF) can be applied to these elements that pass through the ER.

Likewise, downstream of the ER, when salt (with entrained radionuclide impurities) is distilled in the product and MWF distillation furnaces, and then condensed in the salt receivers, the attenuation of the normally nonvolatile radionuclides again increases to 1 and then decreases conservatively to 10^{-3} (0.1 wt% or 1,000 DF). So as long as any radionuclide is in a nominally nonvolatile form at the nominally ambient temperature and pressure conditions of the hot cell, then the DF for evolution of that radionuclide to the cell gas can be conservatively estimated at 1,000. This is expected to apply to the ^{239}Pu-TRU$_{<1y}$ isotopes, any nonvolatile forms of iodine, and any nonvolatile forms of tritium.

As long as the cell gas handling system can achieve an additional DF of at least 10^6 for particulate matter, then the combined DF of up to 10^9 needed for the ^{239}Pu-TRU$_{<1y}$ isotopes can be achieved without any additional emission controls. DFs this high will also achieve needed DFs for nonvolatile forms of iodine and tritium. This overall DF of 10^9 for particulate forms of radionuclides includes, per the adjustment factors in 40 CFR 61 Appendix D:

- DF of 1,000 for radionuclides that are particulate form in the ambient temperature cell gas.
- DF of 1,000 for an initial HEPA prefilter (with a DF of 10) and HEPA filter (with a DF of 100) in the cell gas cleanup system.
- DF of 1,000 for a final HEPA prefilter (with a DF of 10) and HEPA filter (with a DF of 100) before stack discharge.

The adjustment factors in 40 CFR 61 Appendix D are conservative by up to two orders of magnitude, as fabric filters can normally achieve at least 99% control efficiency (100 DF) and HEPA filters can be certified to achieve up to 99.97% efficiency (3,333 DF) (Staley 2004). Assuming the cell gas handling system has at least one HEPA prefilter and one stage of HEPA filtration, then the total conservatively estimated particulate DF can range between 10^9 (using attenuation factors from 40 CFR 61 Appendix D) to 10^{13} (using attenuation factors recommended in Staley 2004). Such high values are difficult to empirically measure but enable regulatory compliance for solid or particulate radionuclides such as would be expected for ^{239}Pu-TRU$_{<1y}$.

7.5 Summary of Strategies for Regulatory Air Emissions Compliance

Table 18 summarizes possible strategies for electrochemical facility compliance to regulatory radionuclide air emissions limits. In all cases, there are reasonably achievable pathways to compliance, although in some cases additional R&D is merited to verify chemical or physical speciation and advance the maturity of potential treatment technologies.

Table 18: Strategies for electrochemical facility compliance to regulatory radionuclide air emissions limits. References to specific DFs are based upon a 200 MTIHM/y electrochemical reprocessing facility as described in the reference case.

Isotope	Estimated overall DFs needed for compliance	Compliance Strategies
^{239}Pu-TRU$_{<1y}$	The fuel cycle-based DF of ~6.1E+9 applies.	The particulate capture performance of the cell gas cleanup system described in Section 7.4 is expected to be able to achieve control of the ^{239}Pu-TRU$_{<1y}$ with DFs ranging from 10^9 to 10^{13}.
^{3}H	Dose-based DFs are ~10. Assuming 70% ^{3}H	Any combination of: 1. Locate the facility in an area of optimal climatological

	loss to the reactor coolant reduces the needed DF to ~3. The DF is proportional to the process rate.	conditions, or in an area of optimal agricultural parameters (nonlocal), to reduce dose to the MEI. 2. Design the facility to achieve better air dispersion to reduce the dose to the MEI – especially by increasing stack height, which may decrease needed dose-based DFs by up to 1–2 orders of magnitude. 3. Design and operate the facility with annual process rates low enough to reduce the ^{3}H dose-based DF to 1. This corresponds to a throughput of 30 MTIHM/y. 4. Demonstrate that catalytic $^{3}H_2$ conversion to condensed tritiated water within the cell gas treatment system meets the needed DF. 5. Demonstrate that a sufficient portion of ^{3}H is retained in less volatile species and not volatilized. 6. Reprocess used fuels with lower burnup or longer cooling times. The amount of tritium in the used fuel decreases approximately proportionately with decreasing burnup. Increasing the cooling time from 2 y by one ^{3}H half-life to 14.5 y would reduce the needed DF by about 50%. 7. Establish the facility boundaries so that the MEI cannot be at the location determined in the air dispersion modeling; such as has occurred at INL due to the distance of INL boundaries from various INL facilities (Sondrup 2020). The farther that an MEI can be located from the facility discharge beyond the location determined in in air dispersion modeling, and in a different direction, the lower the corresponding dose received. 8. Revise the fuel cycle and dose-based DF calculations for less conservatism. For example, increasing the allowed limits from 10% of the regulatory limits to 50% of the regulatory limits decreases the needed DFs by a factor of 5. Considering this suite of options for ^{3}H emissions compliance, no additional ^{3}H-specific control technologies are expected to be required in most scenarios.
^{85}Kr	The fuel cycle-based DF of ~3 applies.	1. Any combination of strategies #1–3 and 6–8 described above as applied to ^{3}H emissions compliance can be used to reduce the needed DFs. Especially consider #8 above: Revise the fuel cycle and dose-based DF calculations for less conservatism. For example, increasing the allowed limits from 10% to 50% of the regulatory limits decreases the needed DFs by a factor of 5. This eliminates the need for any ^{85}Kr control for compliance to fuel cycle limits.

		2. For ^{85}Kr, increase the used fuel cooling time to reduce both the dose-based and fuel cycle-based DFs. Increasing the cooling time from 2 y by one ^{85}Kr half-life to about 12.8 y reduces both DFs by about 50%; increasing the cooling time to about 20 y reduces both DFs by about 67%, reducing the fuel cycle based DF to ~1. 3. Determine if hold-up of ^{85}Kr in the cell gas is a viable way to decay ^{85}Kr to reduce DFs. 4. Capture ^{85}Kr in any cell gas vented to the atmosphere, or in a slipstream of cell gas in the cell gas cleanup system, using such technology options suggested in Section 7.2, with sufficient efficiency to meet DFs.
^{129}I	The fuel cycle-based DF of ~240 applies for process rates below ~ 80 MTIHM/y. Above that process rate, the dose-based DF dominates, and increases with increasing process rate.	1. Any combination of strategies #1–3 and #7–8 as described above for ^{3}H emissions compliance can be used to reduce the needed DFs. Especially consider #8 above: Revise the fuel cycle and dose-based DF calculations for less conservatism. For example, increasing the allowed limits from 10% to 50% of the regulatory limits decreases the needed DFs by a factor of 5. This reduces the fuel cycle-based DF from ~240 to ~50. 2. Demonstrate that iodine speciation in the used fuel and during reprocessing enables an iodine DF greater than the higher of the fuel cycle and dose-based DFs. For example, if it can be demonstrated that over 98% of the iodine is in particulate form (using a fuel cycle based DF of 50), and so is captured with DFs as high as 10^{9} – 10^{13}, then no control is needed for the remaining gaseous iodine species up to 2% of the total iodine. 3. Determine what amounts of gaseous iodine forms exist in the used fuel and during electrochemical reprocessing, and determine how these can be captured with the needed efficiency with a DF on the order of up to ~240 (plus a safety factor to ensure compliance during process variations).

8. APPLICABILITY OF ANALYSIS TO THE DEVELOPMENT OF ADVANCED NUCLEAR TECHNOLOGY

As stated previously, a wide range of nuclear reactor types, fuel types, and cladding types are under development. This report focuses only on electrochemical reprocessing of a metallic SFR U-TRU fuel. However, the analysis methodology used in this report, adapted from the methodology developed for analyzing off-gas control requirements for aqueous reprocessing, can also be used for reprocessing other used fuel types from other reactor types. If this approach is to be translated to other advanced nuclear technologies, some initial thoughts are provided here.

8.1 Processing of Alternative Fuels

The analysis provided here was limited in scope to metallic fuel such as might be used in an SFR. However, the development of advanced nuclear technology includes a wide selection of fuels, including TRISO fuels, oxide based fuels, uranium silicides and uranium nitrides, and molten salts.

8.1.1 Processing of Oxide-based Fuels

Reprocessing of oxide fuels has two notable effects. First, it adds an oxide reduction step before the electrorefining operation. Secondly, it adds a ^{14}C source term to the potential total dose to the public, so (a) the fate of ^{14}C during oxide reduction and electrochemical reprocessing would need to be further evaluated, and (b) air dispersion modeling would be needed to assess if and how much emissions of ^{14}C would need to be controlled. Indications are that carbon is retained in the ER salt through the formation of carbon black on the molten salt (NEA-OECD, 2018). If so, and if the dose from even uncontrolled ^{14}C emissions is as small as was determined for ^{14}C emissions from aqueous used fuel reprocessing (Jubin 2012a), then no isotopic-specific control for ^{14}C emissions may be needed.

8.1.2 Processing of Molten Salts

The processing of molten salt nuclear reactor fuel will give rise to many of the same technological questions as are identified here. The isotopic source term may be different and the processing scheme may be slightly altered, but the behavior of vapor and particulate species will be highly relevant, as they are in this analysis. It is likely that the mitigation strategies discussed here could be easily translated to the reprocessing of molten salt reactor fuels, and even to the operation of molten salt reactors, which have requirements for off-gas treatment of the salt during reactor operation.

8.1.3 Processing of Other Fuels

Reprocessing concepts for the recovery of uranium or plutonium from other fuels (U-silicides, U-nitrides, TRISO fuel, etc.) may involve other reprocessing technologies besides electrochemical reprocessing. The methodology used in this study may be applied to assess off-gas control needs for essentially any used fuel and reprocessing technology.

8.2 Waste Dehalogenation Process

The salt used in electrorefining of used nuclear fuel is recovered and reused for multiple cycles. However, fission product buildup in the salt will eventually lead to treatment to discard the fission products in a suitable waste form. While the CWF described in this report has been developed to dispose fission product-laden salt, alternative waste forms for the fission products have also been studied. These alternatives, including silica–alumina–phosphate (SAP) composite and Fe–P–O (iron phosphate) glass waste forms, have been studied to reduce the amount, or improve the durability, of the fission product waste form, or enable reuse of the chlorine in the salt (Frank 2015, Riley 2020). The release of chlorine from the waste salt is also expected to release iodine. Design for capturing chlorine from salt dehalogenation using phosphate reagents, for chlorine reuse or disposal, and managing co-flowing iodine, has been recently considered. The flowchart in Figure 15 from Soelberg (2020) shows potential decision points and technologies (which are generally not technically mature for these applications) that could be needed to assess management of this chlorine and iodine emissions control. Further evaluation was beyond the scope of this report.

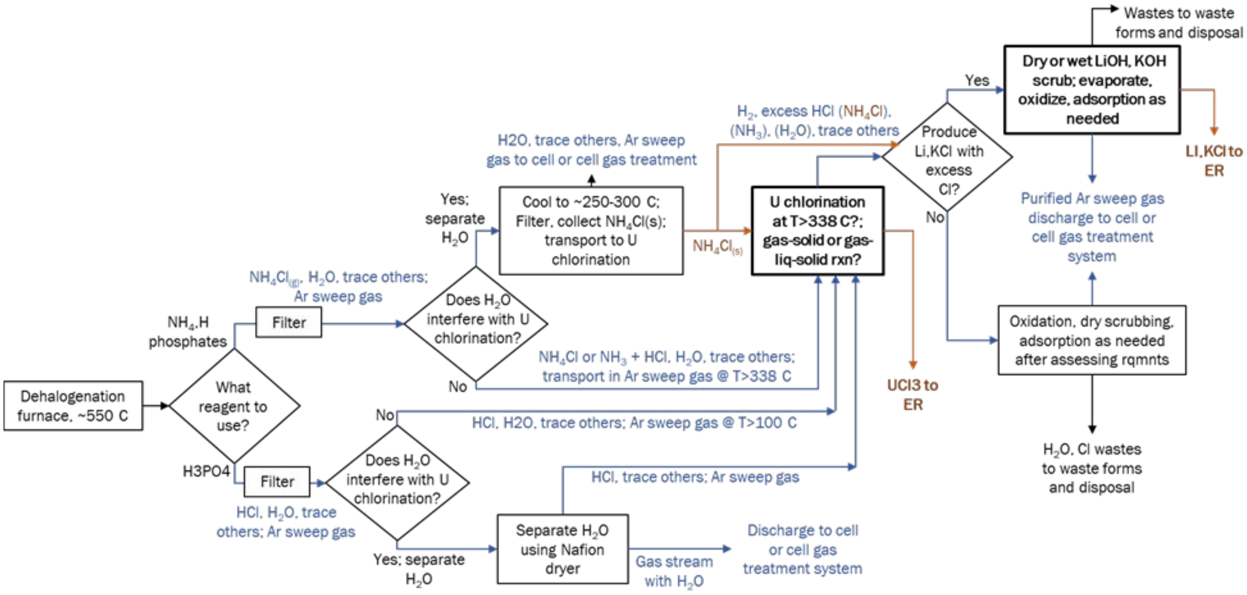

Figure 15: Example flowchart for evaluating the management of chlorine and iodine released from electrochemical salt waste dehalogenation (Soelberg 2020).

8.3 Limitations of Analysis

This analysis was based on a generic electrochemical reprocessing flowsheet and facility design, assuming that all operations are conducted in a argon-blanketed hot cell with continuous purification of cell gas to remove oxygen. If actual implementation varies significantly from this plant design, emissions could vary as well. Two overarching design decisions addressed in the next two sections could influence the extent of mitigation required.

Simplifying assumptions were made in the performance of this study. Different assumptions of (a) fuel type, burnup, and cooling; (b) reprocessing technologies and facilities; (c) how product and waste streams from reprocessing are produced and managed; and (d) application of air emissions regulations and tools such as air dispersion modeling can affect the analysis results.

Although this analysis provides a generic basis for assessing off-gas control needed for electrochemical reprocessing of advanced reactor metal fuels, a site-specific analysis for a real future facility design, guided by the analyses of this study, may have different results.

8.3.1 Characteristics of Irradiated Fuel

Cooling of the fuel after reactor discharge can decrease the mitigation required for the shorter-lived isotopes, including ^{3}H and ^{85}Kr. To achieve these benefits, cooling of 11–12 y would remove ½ of the specific radionuclide (^{85}Kr $t_{1/2}$ = 10.85 y; ^{3}H $t_{1/2}$ = 12.3 y) and decrease off-gas mitigation requirements correspondingly. If a fast reactor fuel cycle at equilibrium can include some postreactor cooling, then this could reduce or eliminate mitigation requirements for tritium and krypton.

The burnup (uranium consumed) of the fuel being processed will obviously affect the radionuclide composition of the spent nuclear fuel. Iodine and krypton scale linearly with burnup. The concentrations of fission products in the used fuel evaluated in this study scale approximately linearly with burnup. The consequence of this is that while compliance to the fuel cycle limits in 40 CFR 190.10(b) is not affected by burnup (because the fuel cycle limits are based on the amounts of ^{85}Kr, ^{129}I, and $^{239}Pu\text{-}TRU_{<1y}$. produced per gigawatt electric [GWe] energy produced), compliance to the dose-based limit in 40 CFR

190.10(a) is affected by burnup. Higher-burnup used fuels may need higher off-gas emissions mitigation to meet the dose-based limit.

Tritium is known to migrate through stainless-steel-clad metal fuels into SFR coolant. When the used stainless-steel clad metal fuels are reprocessed, the amount of remaining tritium could be on the order of only 30% of the amount of tritium produced during irradiation. While this has been accounted for in this study of electrochemical reprocessing of advanced reactor metal fuels, evaluations of tritium emissions from reprocessing other fuel types need to consider how much tritium is retained in those used fuels. Although tritium diffuses through stainless-steel cladding, it does not readily diffuse through zirconium-based cladding. Development of alternative cladding materials is a substantial area of ongoing research, and tritium interactions with alternative cladding materials must be understood.

8.3.2 Emission into a Common Cell Gas

Electrochemical reprocessing at this time is based on batch-type processes that are at least periodically open to the cell atmosphere. This allows radionuclide releases to the cell gas from chopping, electrorefining, melting, and other unit operations. This is a very impactful design decision as it relates to off-gas management. Release into a common cell gas, as opposed to capturing and treating gas streams that evolve from the furnaces, results in the following:

1. Dilution of species to be mitigated. Lower concentrations of gaseous species are typically more difficult to capture.
2. Loss of resolution between unit operations. If a single unit-operation (such as the ER) releases a significant fraction of a selected radionuclide, mitigation of that operation could reduce or eliminate mitigation needs for emissions from other unit operations. It also results in mixtures of chemical species and the potential for interactions of selected radionuclides with cell contents.
3. Dispersion of particulate matter in the hot cell. Species that are volatile or are entrained in furnace gas as particulate matter (fume) at furnace temperatures that can range between 500–1,700°C, can be emitted to the cell atmosphere. Upon cooling to the hot cell ambient temperature, volatilized matter can condense to particles. This or particulate formation (and the size and properties of the resulting particulate and fume) can affect the distribution of selected radionuclides, including iodine and tritium, between particulate and vapor, spread contamination in the hot cell, and affect mitigation strategies.

9. KNOWLEDGE GAPS

Performance of this assessment revealed several areas where information is lacking or additional research is required. These are grouped here into three categories relating to the availability of spent fuel compositions, the release of radionuclides from electrochemical processing operations, and the application of mitigation technologies to the electrochemical processing off-gas.

9.1 Used Fuel Composition

A used fuel composition that has been used in several prior studies (Law 2015, Vienna 2015a, Vienna 2015b) was used for this study. Analyses with a variety of driver and blanket fuels, with different enrichment, burnup, cooling, impurities, and activation products may have provided a wider range of analysis results. However, obtaining custom used fuel compositions for the reference scenario (metallic fuel discharged from a sodium fast reactor) was challenging, and efforts to generate these new data sets was determined to be outside the scope of this report.

Light water reactor modeling and simulation is an active research field and is financially supported by both governmental and industrial partnerships. The suite of tools available to enable researchers outside the field of modeling and simulation to obtain computational data is well developed. One of the premier codes for light water reactor simulation is the SCALE code, which is used for nuclear safety analysis and design. SCALE is maintained by ORNL and funded by the US NRC and the US DOE (Wieselquist et al. 2020). It is explicitly intended to enable an occasional or novice user to conduct simulations. SCALE contains many ready-to-use data sets that can facilitate simulation of light water reactors, but similar libraries are not readily available for SFRs and other reactor types. A review of available resources by the authors indicates that there are active research efforts dedicated to improving modeling and simulation of advanced reactors, but these efforts are not easily accessible to the occasional user outside the field of modeling and simulation.

9.2 Radionuclide Speciation and Thermodynamic Behavior

Tracking radionuclides through the electrochemical processes and performing mass balances to quantitatively determine evolution of radionuclides into gas streams requires understanding their chemical behavior, sometimes with high resolution. For example, release amounts of 10% or less of the tritium inventory or 0.5% or less of the iodine inventory can impact regulatory compliance. This level of fidelity cannot be achieved with existing modeling tools or with the experimental data currently available.

The following specific data needs include, but are not limited to:

- Iodine and tritium mass balances and speciation during different parts of electrochemical reprocessing. Partitioning between particulate (solid-phase), dissolved salt-phase, metal phase, and gaseous species is of special interest. This includes:
    - Identifying thermodynamically favored and empirically determined speciation of iodine and tritium during electrochemical reprocessing unit operations.
    - Identifying partial pressures of iodine species such as NaI, CsI, and I_2 above a mixture dominated by bond Na.
    - Identifying partial pressures of relevant iodine and tritium species above a chloride-based salt melt and during such processes as vacuum distillation and metal melting.
    - Determining the fate of less-volatile radionuclide species after evolution from electrochemical reprocessing equipment to the cell gas, including the formation of particles, particle size, and particle deposition in the hot cell.
- Behavior of gaseous iodine forms in the atmosphere, if released to the atmosphere during reprocessing. Can gaseous iodine forms pass through the cell gas treatment system to the atmosphere, and then convert in the atmosphere to particulate forms, which cause higher doses to the public?

In addition to questions of chemical speciation, questions about the physical form of iodine emissions are very important to the scenario under examination. Particulate iodine emissions can be easier to mitigate than vapor-phase species. The potential iodine emissions from electrochemical reprocessing operations may include a mixture of vapor phase species and particulate matter entrained from the furnaces into the cell gas. Vapor phase species that are emitted directly into the inert cell may interact to form particulate, or they may remain vapor. However, if vapor phase iodine is released to the atmosphere, and then undergoes subsequent reactions with atmospheric gases or aerosols after their release to form particulate phases, these can result in relatively high doses to the MEI. A better understanding of the complexities related to iodine physical form would allow for a substantially more precise analysis of needed mitigation.

9.3 Maturity of Mitigation Technology

Capture of ^{85}Kr, ^{3}H, and ^{129}I released into the cell off-gas may be needed depending on conditions of electrochemical reprocessing addressed in this report. Some aspects of control technologies proposed for these isotopes in argon cell gas cleanup systems are based on limited information or immature technologies for this application. Further information and technology maturation is needed in the following areas before specific mitigation technologies can be deployed.

9.3.1 Noble Gas Mitigation

Cryogenic capture of noble gases was included in the design of the commercial-size IFR fuel cycle facility (Lineberry 1985). Since then adsorption of noble gases on sorbents at temperatures higher than cryogenic temperatures has been studied, although primarily for noble gas capture in aqueous reprocessing off-gas streams as is summarized in Section 7. Neither cryogenic noble gas capture nor adsorption on sorbents has been proven at full scale for noble gas capture in argon cell gas streams for electrochemical reprocessing. The behavior of noble gas sorbents within the conditions expected in the cell gas treatment system has not yet been examined. The effects of argon as the balance gas on the noble gas separation should be studied. It is not known how exposure to residual chlorine or iodine may affect the adsorption behavior or sorbent structure. Testing of noble gas separation in prototypic gas streams is recommended.

9.3.2 Removal of ^{3}H

Tritium control has been proposed using the catalytic oxidation and moisture adsorption process used to remove oxygen in argon cell cleanup systems. The performance and effectiveness of this process for tritium control has yet to be demonstrated. The effectiveness in capturing tritium may depend on the speciation of tritium between H_2 and hydrogen halide, as well as on the stoichiometry needed to achieve both oxygen and tritium control. Chemisorption of halogen gases on the catalyst may interfere with the oxidation processes, or this chemisorption may be a way to also achieve iodine control in the same process by making the catalyst a consumable iodine sorbent that is replaced as needed. This could be a way to achieve cell gas oxygen, tritium, and iodine control all with one process. Options like these, circumstances such as catalyst operating temperatures, and potential levels of chlorine species at concentrations higher than iodine species, require further technology development and demonstration.

Adsorption of tritium may also proceed by adsorption by molecular sieves, if tritium is in the form of polar molecules such as HCl and HI. HCl adsorption on molecular sieves is not well understood at low partial pressures of HCl. Separation factors for HCl/Cl_2 using molecular sieves at the expected concentrations should be collected. Capture of iodine, if in the form of HI, could be an added (but as yet unproven) benefit of this technology. The performance of this technology requires further development and demonstration for this application.

9.3.3 Iodine Speciation and Adsorption

Chemisorption of iodine gas species on silver-based sorbents and other sorbents has been studied for decades and has most recently been advanced for iodine capture in aqueous reprocessing off-gas streams. Iodine chemisorption on silver-based sorbents is a viable technology for use in the treatment of off-gas streams arising from electrochemical reprocessing, but it needs to be demonstrated for use in prototypic gas streams. For example, iodine adsorption dynamics, sorbent capacity, and co-sorption of chlorine gas species need to be evaluated. Alternative iodine sorbents that do not contain silver are limited in number and have not been demonstrated in representative applications. Identification and development of alternative iodine sorbents should be prioritized.

10. CONCLUSIONS

An assessment has been conducted to determine how key regulations regarding volatile radionuclide emissions to the atmosphere may apply to the off-gas streams associated with electrochemical reprocessing. The scope of this assessment was based upon a generic electrochemical reprocessing scheme with a throughput rate of 200 MTIHM/y applied to metallic fuel discharged from an SFR, but the findings are able to be translated to other advanced nuclear scenarios as merited. A reference spent fuel composition with an initial composition of 70wt%U–20 wt% TRU–10 wt% Zr, a burnup of 99.6 GWd/MTIHM, and a cooling time of 2 y was used as an input to air dispersion modeling. The equilibrium composition of the TRU components of the used fuel for the system with a conversion ratio of 0.75 is 0.21% Np–17.7 wt% Pu–0.92 wt% Am–0.38 wt% Cm.

10.1 Decontamination Factor Estimates

Air dispersion modeling was performed using the CAP-88 model and evaluated using the facility-wide DFs that would be required to achieve regulatory compliance with the dose-based limits set forth by 40 CFR 190.10(a), assuming that this reprocessing facility would be an NRC-licensed facility and not a DOE facility, which would have different regulatory limits. These DFs were compared to those required by fuel cycle–based limits set forth by 40 CFR 190.10(b). The required DFs were calculated assuming full release of the selected radionuclides from the facility and that each radionuclide could not contribute more than 10% to the regulatory limits. The dose to the maximum exposed individual was used in determining the DF for compliance to the dose-based limit. Two theoretical sites with disparate climatological conditions were selected for air dispersion modeling (Idaho and Tennessee). The theoretical facilities were assumed to have a stack height of 50 m (164 ft), a stack diameter of 2.2 m; and a momentum-type plume with a stack exit velocity of 12.2 m/s (that corresponds to volumetric rate of 46 m^3/s or 98,000 scfm).

The radionuclides modeled included ^{3}H, ^{85}Kr, ^{129}I, and ^{239}Pu-TRU$_{<1y}$. It was found that 40 CFR 190.10(b) is the most restrictive regulation for ^{85}Kr and ^{239}Pu-TRU$_{<1y}$, with DFs of 3 and 6.1E+09, respectively. 40 CFR 190.10(a) could require mitigation of tritium in some scenarios, with an estimated DF of about 3 for the reference scenarios. The tritium DF varies proportionately with the used fuel process rate.

The fuel cycle-based limit in 40 CFR 190.10(b) for ^{129}I results in a DF of about 240 for iodine. The need for iodine mitigation based on dose to the public per 40 CFR 190.10(a) depended upon the physical form of iodine as either particulate or vapor-phase species and was also different than the fuel cycle-based limit. Emission of iodine from the facility as a vapor necessitated dose-based DFs of 2 but emission as a particulate would require DFs of >6,000 to meet thyroid dose-based limits of 40 CFR190.10(a) for a 200 MTIHM/y used fuel reprocessing rate. The effects of physical form on needed iodine mitigation are significant, but the understanding of whether iodine will exist as vapor or particulate after emission is limited. Particulate-phase iodine is easily mitigated within the facility by standard HEPA filtration, but vapor-phase iodine could require additional capture operations. Understanding the physical form of iodine both within the electrochemical processing operations and after emission from the facility stack was identified as key to understanding the DFs that may be required in the operation of an electrochemical reprocessing facility and is recommended as an area important for additional study.

Whether or not additional off-gas controls (beyond common operations such as HEPA filtration and oxygen and moisture control) are needed for any of these regulated or volatile radionuclides depends on the (a) type of facility (NRC-regulated or DOE), (b) used fuel process rate, (c) used fuel burnup and composition, (d) speciation and retention of volatile radionuclides in the process and in the cell gas cleanup system, (e) site-specific parameters such as location, meteorology, stack height, and site boundaries, and (f) levels of conservatism and safety factors used in assessing compliance to air emissions regulations.

10.2 Speciation and Partitioning during Electrochemical Reprocessing

After generation of facility-wide DFs, the electrochemical processing unit operations were evaluated to identify potential release points for the volatile radionuclides and to assess the potential for retention of the radionuclides within the process (thus decreasing the need for mitigation). Notably, no retention of noble gases is expected in the process due to high-temperature (>500°C) fuel dissolution in the salt melt.

While quantitative speciation of tritium to the vapor phase compounds such as H_2 and HCl might be expected, the potential for speciation to less volatile species such as metal hydrides (and the distribution between different gaseous species) is not well known.

Limited data exists to support estimation of iodine release from the electrochemical reprocessing operations. While it is likely that iodine solubility and retention in the ER salt is high, even a loss of 0.5% to the cell gas in the form of volatile iodides could prevent regulatory compliance without added iodine capture in the cell gas cleanup system. No experimental data exists with this level of precision.

10.3 Potential Mitigation Technologies

Mitigation technologies for ^{3}H, ^{85}Kr, ^{129}I, and ^{239}Pu-TRU$_{<1y}$ were identified. In all cases, there are reasonably achievable pathways to compliance, although in some cases additional R&D is merited to verify the chemical speciation of these isotopes and the development and demonstration of potential treatment technologies for this application. A key assumption within this assessment was that emissions from the processing operations are released directly to a common argon-filled hot cell. This results in dilution of the radionuclides and can complicate removal. However, the argon cell gas is also continuously cleaned and recycled, and this process can influence the mitigation of the selected radionuclides. Notably, ^{239}Pu-TRU$_{<1y}$, assumed to be fully particulate and refractory in nature, can be removed using standard HEPA filtration with sufficiently high efficiencies. Any iodine emitted from the unit operations as particulate will be similarly filtered.

The behavior of ^{3}H within the cell-gas treatment system is not fully understood, specifically its conversion to water within the O_2 catalytic removal operation and subsequent capture in regenerable molecular sieve absorbents. If ^{85}Kr control is needed, potential capture technologies include cryogenic distillation and absorption by solid sorbents (including engineered zeolites and metal organic frameworks) at temperatures higher than cryogenic. If desired, both cryogenic distillation and absorption by solid sorbents can be configured to separate Kr from non-radioactive Xe, decreasing the waste volumes generated by the disposal of Xe with Kr.

Iodine also has the potential to interact with the catalytic oxygen removal operation within the cell-gas treatment train, potentially consuming the catalyst. If emitted as vapor, iodine and chlorine may be chemisorbed on the oxidizer catalyst. Metals like silver in the oxidizer catalyst or in separate silver-based sorbents can chemisorb the halogens and remove iodine effectively, but consumption of the catalyst or sorbent may be high enough to drive interest in alternative iodine sorbents for this application.

10.4 Recommendations for Future Study

Performance of this assessment revealed several areas that information is lacking or additional research is required in order to better determine if or what kinds of off-gas control might be needed for actual electrochemical reprocessing facilities. First, and most significantly, the understanding of the chemical speciation and physical form and partitioning of iodine during electrochemical processing operations is lacking and prevents the ability to accurately assess the potential iodine mitigation requirements. Future research in this area should be multifaceted and include thermodynamic modeling of iodine speciation in different process steps, experiments to quantify the kinetics of vapor-phase and melt-phase transitions, bench-scale experiments to determine the potential chemical and physical form of iodine emissions from the ER, and verification of iodine behavior with experiments utilizing operational facilities. Similarly, an improved understanding of iodine behavior in the environment after release from the facility stack will be required to refine dose estimations, as particulate and vapor-phase emissions result in significantly different doses to the MEI.

In the mitigation of specific radionuclides, several key topics merit additional R&D. First, the verification of tritium behavior within a conceptual cell gas treatment system should be performed. Second, the development of alternative iodine sorbents (those not including hazardous or precious metals) should be pursued. Third, solid sorbents previously developed for noble gas separations should be tested in conditions that might be expected in this application.

The assessment conducted here provides a useful reference case for determining if and how off-gas emissions controls might be needed for electrochemical reprocessing of advanced reactor metallic fuels. The methodology used here can also be applied to other advanced nuclear technologies, including the operation of molten salt reactors and the reprocessing of advanced nuclear fuels. Importantly, the knowledge gaps identified as relating to the specific scenario evaluated (electrochemical reprocessing of metallic SFR fuel) will also be applicable to other advanced nuclear scenarios too, allowing for R&D to support multiple technologies. Future research should focus on fundamental chemistry of volatile radionuclide release from electrochemical processing and subsequent adsorption of volatile radionuclides so that as additional nuclear technologies approach deployment the scientific barriers to implementation will be reduced.

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BIBLIOGRAPHY

Ackerman, J.P., T.R. Johnson, L.S.H. Chow, E.L. Carls, W.H. Hannum, and J.J. Laidler, 1997, “Treatment of Wastes in the IFR Fuel Cycle,” *Progress in Nuclear Energy*, 31(1/2): 141–154.

Allen, J. (2018, November 13). GE Hitachi and PRISM Selected for U.S. Department of Energy's Versatile Test Reactor Program. Retrieved August 24, 2020, from https://www.ge.com/news/press-releases/ge-hitachi-and-prism-selected-us-department-energys-versatile-test-reactor-program

Baum, E.M., H.D. Knox, and T.R. Miller, 2002, *Nuclides and Isotopes: Chart of the Nuclides*. Schenectady, NY: Knolls Atomic Power Laboratory.

Berl, W. G., Ed., *Physical Methods in Chemical Analysis: Vol. IV*, Academic Press, New York, 1961.

Bruffey, S.H., R.T. Jubin, D.M. Strachan, N.R. Soelberg, B.B. Spencer, and B.J. Riley, 2020, Performance Criteria for Capture and/or Immobilization Technologies, Revision 1, ORNL/SPR-2020/1583, Oak Ridge National Laboratory, Oak Ridge, TN, June.

Cahalan, J., T. Fanning, M. Farmer, C. Grandy, E. Jin, T. Kim, and Y. Momozaki, 2007, Advanced Burner Reactor 1000MWth Reference Concept, No. ANL-AFCI-202, Argonne National Lab, Lemont, IL.

CAP-88 PC. (2020, May 08). Retrieved August 31, 2020, from https://www.epa.gov/radiation/cap-88-pc

Choi, J.H., I.H. Cho, H.C. Eun, H.S. Park, Y.Z. Cho, K.R. Lee, G.I. Park, S.H. Kim, C.H. Shin, and J.K. Kim, 2014, “Fabrication and Physical Properties of Lanthanide Oxide Glass Wasteform for the Immobilization of Lanthanide Oxide Wastes Generated from Pyrochemical Process,” *Journal of Radioanalytical and Nuclear Chemistry*, 299: 1731–1738. DOI: 10.1007/s10967-013-2863-6.

Dixon, B., G. Matthern, N. Soelberg, T.K. Kim, and M. Todosow, 2019, Technology Maturation Planning and Technology Roadmap Development Example Using the Technology and System Readiness Assessment Process, INL/LTD-19-54549, Idaho National Laboratory, Idaho Falls, ID, June 28.

Department of Energy (DOE), 2007, "Draft 30% Conceptual Design Report for the Advanced Fuel Cycle Facility," Section 3.5, Pyrochemical Separations, prepared for the Department of Energy Office of Nuclear Energy Global Nuclear Energy Partnership Program, January 26.

Ebersole, E.R., W.R. Vroman, and J.R. Krsul, 1971, "Tritium in the EBR-11 Reactor Complex," Argonne National Laboratory – Idaho, *Transactions of the American Nuclear Society*, 1971 Annual Meeting, Boston, MA, June 13–17.

Eng, J. and S. H. J. Greenwood, 1961, "Chlorine Separation Process," US Patent 3,001,607, Sept. 26.

Environmental Protection Agency (EPA). 1999. EPA 402-R-99-001 Federal Guidance Report 13. "Cancer Risk Coefficients for Environmental Exposure to Radionuclides". USEPA Office of Radiation and Indoor Air, Washington, DC.

Environmental Protection Agency (EPA). 2010a. Chapter 40. Environmental Protection Agency: Part 61–National Emission Standards for Hazardous Air Pollutants. Subpart H—National Emission Standards for Emissions of Radionuclides Other Than Radon from Department of Energy Facilities, 92—Standard. 40 CFR 61.92. US Environmental Protection Agency, Washington, DC.

Environmental Protection Agency (EPA). 2010b. Protection of Environment: Chapter I–Environmental Protection Agency (Continued), Part 190 Environmental Radiation Protection Standards for Nuclear Power Operations. 40 CFR 190.10. US Environmental Protection Agency, Washington, DC.

Frank, S.M, 2011a. Development and Characterization of New High-Level Waste Forms for Achieving Waste Minimization from Pyroprocessing, I-NERI-2007-004-K, INL/EXT-11-23352, Idaho National Laboratory, Idaho Falls, ID.

Frank, S.M. 2011b. Radiogenic Iodine Partitioning during the Electrochemical/Pyrometallurgical Treatment of Na-bonded Metallic Fuel, FCRD-WAST-2011-000091, September.

Frank, S. 2014. "E.1.e.2. Iodine Capture from LiCl, Section 3.11," CRADA contribution of iodine off-gas.

Frank, S., W. Ebert, B. Riley, H.S. Park, Y.Z. Cho, C.H. Lee, M.K. Jeon, J.H. Yang, and H.C. Eun, 2015, Waste Stream Treatment and Waste Form Fabrication for Pyroprocessing of Used Nuclear Fuel, INL/EXT-14-34014, Idaho National Laboratory, Idaho Falls, ID, July.

Fast Reactor Working Group (FRWG), 2018, "Nuclear Metal Fuel: Characteristics, Design, Manufacturing, Testing, and Operating History," White Paper 18-01, Fast Reactor Working Group (FRWG), June 2018.

Goff, K.M., J.C. Wass, K.C. Marsden, and G.M. Teske, 2011, "Electrochemical Processing of Used Nuclear Fuel." *Nuclear Engineering and Technology*, 43(4): 335–342.

Huve, J. et al. 2018. "Porous sorbents for the capture of radioactive iodine compounds: a review." *RSC Advances* 8(51): 29248–29273.

Idaho Department of Environmental Quality, 2020. 58.01.01 – Rules for the Control of Air Pollution in Idaho. Idaho Department of Environmental Quality Air Quality Divisions. https://adminrules.idaho.gov/rules/current/58/580101.pdf.

International Atomic Energy Agency (IAEA), 2012. Liquid Metal Coolants for Fast Reactors (Reactors Cooled by Sodium, Lead and Lead-bismuth Eutectic). International Atomic Energy Agency, Vienna Austria.

International Commission on Radiological Protection (ICRP), 1996. "Age Dependent Doses to Members of the Public from Intake of Radionuclides, Part 5. Compilation of Ingestion and Inhalation Dose Coefficients" ICRP Publication 72, Pergamon Press, Oxford.

Jubin, R. T. 1979. A literature survey of methods to remove iodine from off-gas streams using solid sorbents. Department of Energy, Oak Ridge National Laboratory, Chemical Technology Division.

Jubin, R.T., N. Soelberg, and D.M. Strachan, 2012b, "Position Paper on Practicable Performance Criteria for the Removal Efficiency of Volatile Radionuclides," Web. doi:10.2172/1046075.

Jubin, R.T., Jordan, J.A., Spencer, B.B., Soelberg, N.R., Welty, A.K., Greenhalgh, M., Strachan, D.M., Thallapally, P.K. 2016a. Engineering Evaluation of an Integrated Off-Gas Treatment System for Used Nuclear Fuel Reprocessing Facilities. FCRD-MRWFD-2016-000313, Oak Ridge National Laboratory, Oak Ridge, TN.

Jubin, R.T, N.R. Soelberg, D.M. Strachan, and G. Ilas, 2012a, "Fuel Age Impacts on Gaseous Fission Product Capture During Separations," FCRD-SWF-2012-000089, September 21.

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

Jubin, R.T., D.M. Strachan, G. Ilas, B.B. Spencer, and N.R. Soelberg, 2014, "Radioactive Semivolatiles in Nuclear Fuel Reprocessing," FCRD-SWF-2014-000601, ORNL/LTR-2014/354, INL/EXT-14-33122, , Oak Ridge National Laboratory, Oak Ridge, TN, September.

LaPlace, A., J. Lacquement, J.L. Willit, R.A. Finch, R.A., G.A. Fletcher, and M.A. Williamson, 2008, "Electrodeposition of Uranium and Transuranic Metals on Solid Cathodes," *Nuclear Technology*, 163(3): 366–372.

Law, J., N. Soelberg, T. Todd, G. Fredrickson, S. Frank, C. Pereira, M. Williamson, W. Ebert, J. Willit, R. Jubin, J. Vienna, G. Lumetta, J. Crum, T. Rudisill, J. Bresee, C. Phillips, B. Willis, and S. Bader, 2015, Separation and Waste Form Campaign Full Recycle Case Study, FCRD-MRWFD-2015-000241, September 30.

Lee, C., D. Kim, and Y. Jung, 2001, "Fission Gas Release and Swelling Model of Metallic Fast Reactor Fuel," *Journal of Nuclear Materials*, 288: 29–42.

Lee, C.B., J.S. Cheon, S.H. Kim, J.-Y. Park, and H.-K. Joo, 2016, "Metal Fuel Development and Verification for Prototype Generation IV Sodium-Cooled Fast Reactor," *Nuclear Engineering and Technology*, 18: 1096–1108.

Lineberry, M.J., R. D. Phillips, and J.P. Burelbach, 1985, Commercial-Size IFR Fuel Cycle Facility: Conceptual Design and Cost Estimate, ANL-IRF-25, Argonne National Laboratory, Lemont, IL, October.

Marsden, K. (Idaho National Laboratory), and S. Frank (Idaho National Laboratory), 2020, personal communication to N. R. Soelberg (Idaho National Laboratory), April 13.

Morrison, M.C. and K.J. Bateman, 2010, Perform 2nd Pre-Qualification CWF Test Run, FCRD-SEPA-2010-000057, Idaho National Laboratory, Idaho Falls, ID.

Morrison, M.C., K.J. Bateman, and M.F. Simpson, 2010, "Scale up of Ceramic Waste Forms for the EBR-II Spent Fuel Treatment Process," 3rd International Pyroprocess Research Conference, Research Institute of Atomic Reactors, Dimitrovgrad, Russia.

Nandanwar, S.U., J. Dantas, K. Coldsnow, M. Green, V. Utgikar, P. Sabharwall, and D.E. Aston, 2016, "Porous Microsphere of Magnesium Oxide as an Effective Sorbent for Removal of Volatile Iodine from Off-gas Stream," *Adsorption*, 22(3): 335–345.

NEA-OECD, *State-of-the-Art Report on the Progress of Nuclear Fuel Cycle Chemistry*, NEA No. 7267, Nuclear Energy Agency, Organisation for Economic Co-operation and Development, 2018.

Nuclear Regulatory Commission (NRC), 2012, Chapter 10, "Energy: Part 20—Standards for Protection against Radiation," 10 CFR 20. US Nuclear Regulatory Commission, Washington, DC.

Pereira, C., M.C. Hash, M.A. Lewis, M.K. Richmann, and J. Basco, 1999, "Incorporation of Radionuclides from the Electrometallurgical Treatment of Spent Fuel into a Ceramic Waste Form," Scientific Basis for Nuclear Waste Management XXII, 556: 115–120, American Ceramic Society, Warrendale, PA.

Priebe, S. and K. Bateman, 2008, "The Ceramic Waste Form Process at Idaho National Laboratory," *Nuclear Technology*, 162: 199–207.

Priebe, S., 2007, "The Ceramic Waste Form Process at Idaho National Laboratory," 26th International Conference on Incineration and Thermal Treatment Technologies, 763–775, Air and Waste Management Association, Phoenix, AZ.

Riley, B.J., J.A. Peterson, J.D. Vienna, W.L. Ebert, and S.M. Frank, 2019, "Dehalogenation of Electrochemical Processing Salt Simulants with Ammonium Phosphates and Immobilization of Salt Cations in an Iron Phosphate Glass Waste Form," *Journal of Nuclear Materials*, 529: 151949, https://doi.org/10.1016/j.jnucmat.2019.151949.

Shigetaka M. and Takafumi A., 2002, "Decay Heat of Fast Reactor Spent Fuel," *Journal of Nuclear Science and Technology*, 39: 1104.

Sigma Aldrich, 2020, personal communication to N. Soelberg (Idaho National Laboratory), July 10.

Simpson, M.F., 2013, "Projected Salt Waste Production from a Commercial Pyroprocessing Facility," *Science and Technology of Nuclear Installations*, Article 945858, http://dx.doi.org/10.1155/2013/945858.

Simpson, M.F., and P. Sachdev, 2008, "Development of Electrorefiner Waste Salt Disposal Process for the EBR-II Spent Fuel Treatment Project," *Nuclear Engineering and Technology*, 40(3): 175–182.

Simpson, M.F., T.S. Yoo, R.W. Benedict, S. Phongikaroon, S. Frank, P. Sachdev, and K. Hartman, 2007, "Strategic Minimization of High Level Waste from Pyroprocessing of Spent Nuclear Fuel," Global 2007, American Nuclear Society, LaGrange Park, IL.

Soelberg, N.R., T.G. Garn, M.R. Greenhalgh, J.D. Law, R. Jubin, D.M. Strachan, and P.K. Thallapally, 2013, "Radioactive Iodine and Krypton Control for Nuclear Fuel Reprocessing Facilities," Hindawi

Publishing Corporation, *Science and Technology of Nuclear Installations*, Article ID 702496, INL/JOU-13-28481, February 19, http://dx.doi.org/10.1155/2013/702496.

Soelberg, N., M. Abbott, D. Haefner, and R. Jubin, “Gaseous Fission Product Emissions Control During Spent Nuclear Fuel Recycling,” 235th American Chemical Society National Meeting and Exposition, New Orleans, LA, April 6–10, 2008.

Sondrup, A. Jeffrey, 2020. “Idaho National Laboratory CY-2018 National Emission Standards for Hazardous Air Pollutants Analysis, Methodology and Results for Radionuclides,” Idaho National Laboratory, May 27, 2020.

Spoerke, E., J. Wheeler, J. Ihlefeld, M. Blea, H. Brown-Shaklee, and M. Rodriguez, 2014, Ion Selective Ceramics for Waste Separations: Performance and Design of Ion Selective Ceramic Membrane for Molten Salt Recycle, FCRD-SWF-2014-000033, Sandia National Laboratories, Albuquerque, NM.

Spoerke, E., J. Wheeler, J. Ihlefeld, M. Blea, K. Waldrip, and L. Johnson, 2013, Ion Selective Ceramics for Waste Separations: End of Fiscal Year Project Assessment, FCRD-SWF-2013-000300, Sandia National Laboratories, Albuquerque, NM.

Staley, C.S., M.L. Abbott, and P.D. Ritter, 2004, INEEL Air Modeling Protocol, INEEL/EXT-04-02511, December.

Vienna, J.D., N.R. Soelberg, W.L. Ebert, J. Willit, M.A. Williamson, S.M. Frank, G. Fredrickson, and B.J. Riley, 2015a. “Initial Salt Waste Management Trade Study,” FCRD-MRWFD-2015-000469, INL/LTD-15-34743, Rev. 0, March 31, 2015.

Vienna, J.D., E.D. Collins, J.V. Crum, W.L. Ebert, S.M. Frank, T.G. Garn, D. Gombert, R. Jones, R.T. Jubin, V.C. Maio, J.C. Marra, J. Matyas, T.M. Nenoff, B.J. Riley, G.J. Sevigny, N.R. Soelberg, D.M. Strachan, P.K. Thallapally, J.H. Westsik, 2015b. "Closed Fuel Cycle Waste Treatment Strategy," FCRD-MRWFD-2015-000674, Rev. B, Pacific Northwest National Laboratory, Richland WA, February 2015.

Volkovich, V.A., T.R. Griffiths, and R.C. Thied, 2003, “Treatment of Molten Saltwastes by Phosphate Precipitation: Removal of Fission Product Elements after Pyrochemical Reprocessing of Spent Nuclear Fuels in Chloride Melts,” *Journal of Nuclear Materials*, 323(1): 49–56.

W. A. Wieselquist, R. A. Lefebvre, and M. A. Jessee, Eds., SCALE Code System, ORNL/TM-2005/39, Version 6.2.4, Oak Ridge National Laboratory, Oak Ridge, TN (2020).

Wigeland, R. et al., 2014, Nuclear Fuel Cycle Evaluation and Screening—Final Report, FCRD-FCO-2014-000106, INL/EXT-14-31465, Idaho National Laboratory, Idaho Falls, ID.

Willit, J.L. and M.A. Williamson, 2012, “A High-Throughput Electrorefiner for Recovery of U and U/TRU Product from Spent Fuel,” US Patent 8,097,142, January 17.

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APPENDIX A: Additional CAP-88 Findings

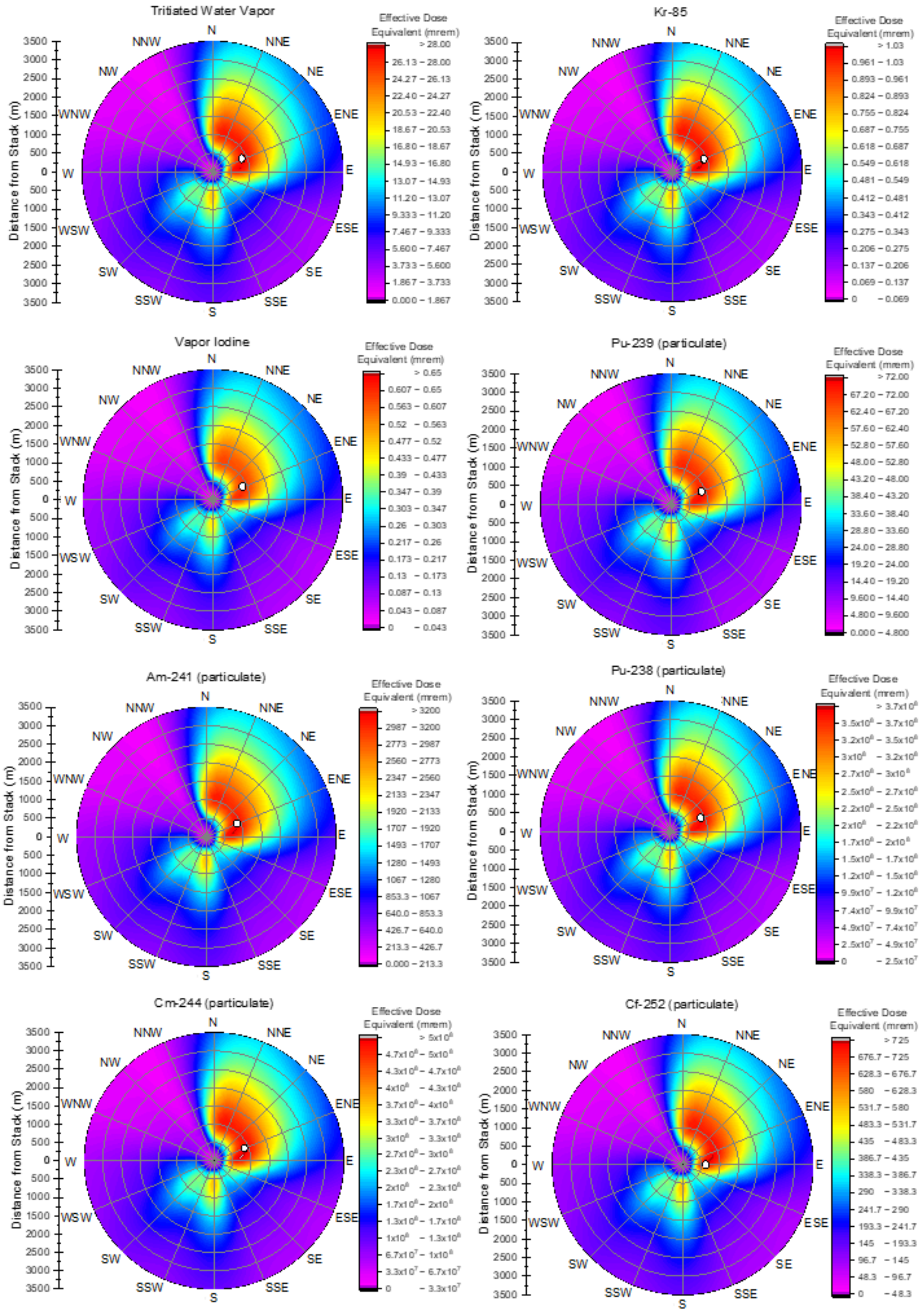

Figure A.1 Heat maps of effective dose equivalent from base case (200 MTIHM/y throughput, 50 m stack height, 12.2 m/s stack gas velocity, 2.2 m stack diameter) as function of direction and distance from stack release in Idaho. White circles represent location of predicted MEI.

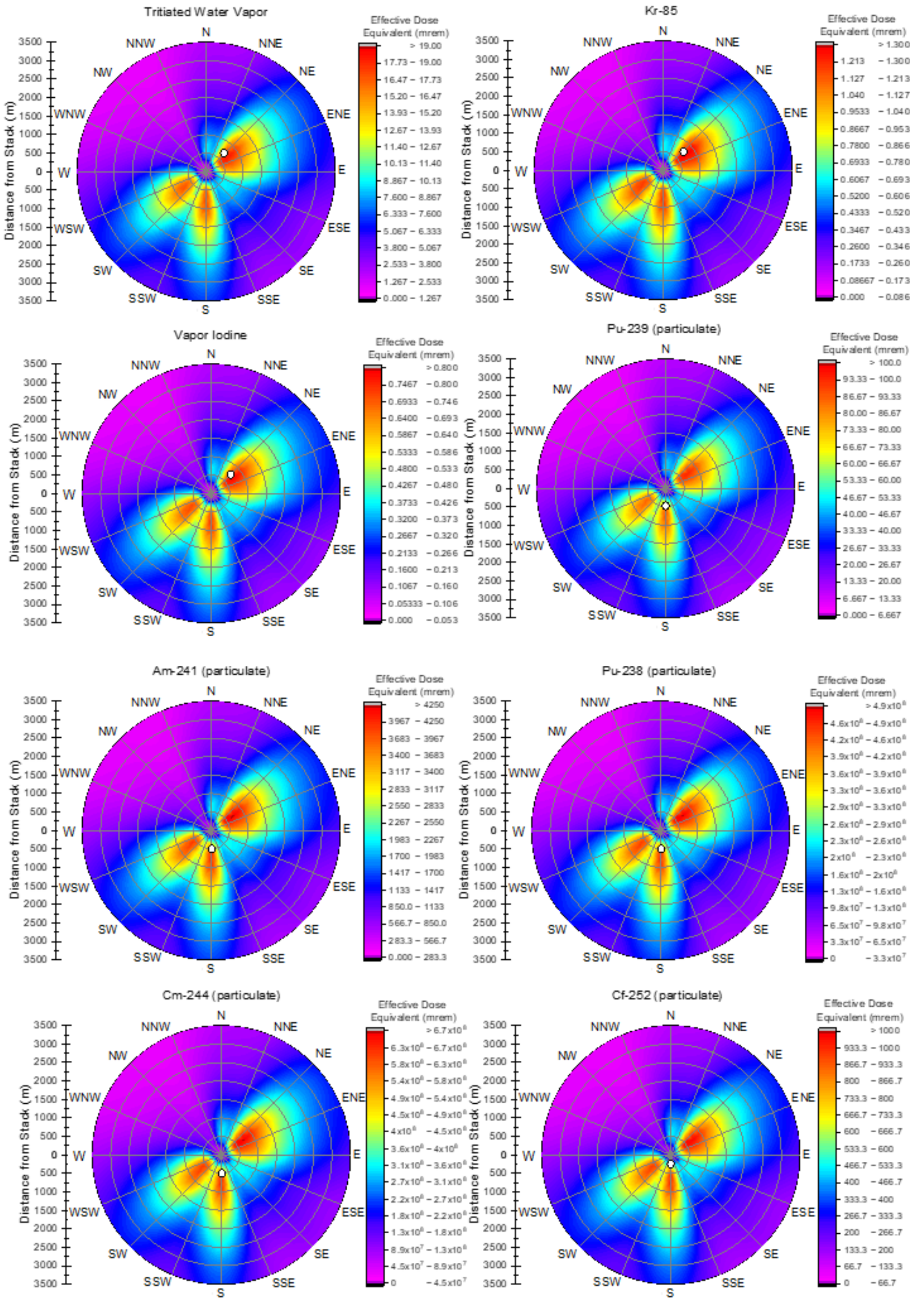

Figure A.2: Heat maps of effective dose equivalent from base case (200 MTIHM/y throughput, 50 m stack height, 12.2 m/s stack gas velocity, 2.2 m stack diameter) as function of direction and distance from stack release in Tennessee. White circles represent location of predicted MEI.

Using radial coordinate plots (Figures A.3 and A.4) as was done for the base case, differences in effective dose equivalent as a function of iodine chemical form can be described in more detail. For the Idaho facility, all chemical forms have similar directions of the MEI from the stack, but the MEI is closer to the stack for particulate iodine forms (Figure A.3). For the Tennessee facility, the particulate iodine forms have an MEI in a nearly opposite direction to the vapor phase and the MEI is significantly closer to the stack for particulate iodine forms (Figure A.4). For both locations, dose maps are similar for both vapor and particulate phases, though the vapor phase appears to be dispersed to farther distances from the stack than particulate, which is not unexpected. Overall, the most notable effect of differing iodine chemical and physical form is observed in the magnitudes of the of effective dose equivalent and DF.

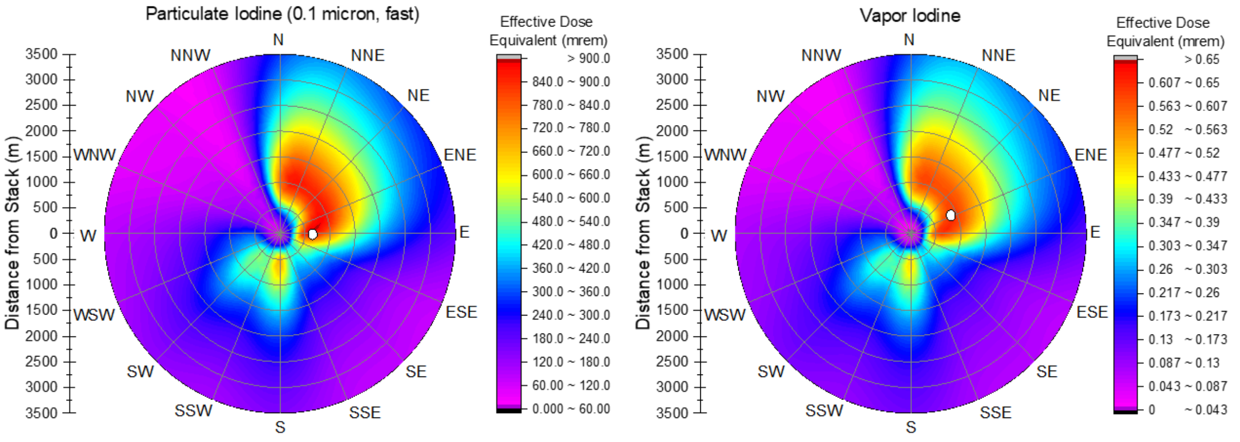

Figure A.3: Heat map of effective dose equivalent from iodine species as function of direction and distance from stack release in Idaho. White circles represent location of predicted MEI. Scale of effective dose equivalent for particulate ranges from 0 to 900 mrem, but scale for vapor ranges from 0 to 0.65 mrem.

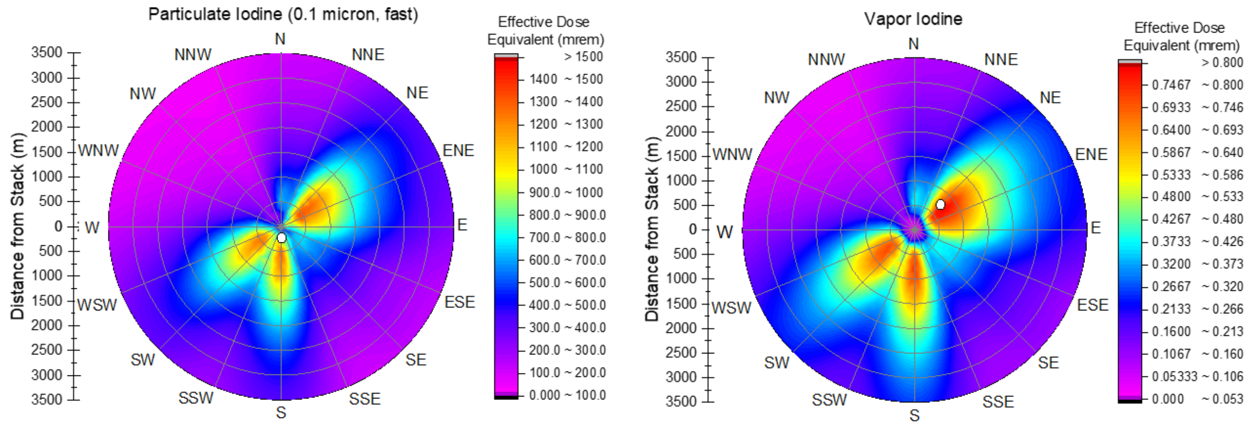

Figure A.4: Heat map of effective dose equivalent from iodine species as function of direction and distance from stack release in Tennessee. White circles represent location of predicted MEI. Scale of effective dose equivalent for particulate ranges from 0 to 1500 mrem, but scale for vapor ranges from 0 to 0.8 mrem.

Table A1: Impact of stack height on dose location and required DF for a facility located in Idaho.

Isotope	Stack Height (m)	Location of MEI	Effective Dose Equivalent (mrem)	DF (10%)
H-3	30	500 m ENE	73.9	3.0E+01
Kr-85	30	500 m ENE	2.71	1.1E+00
I-129	30	500 m ENE	1.61	6.4E-01
Pu-239	30	500 m ENE	186	7.4E+01
Am-241	30	500 m ENE	8420	3.4E+03
H-3	50	900 m ENE	27.4	1.1E+01
Kr-85	50	900 m ENE	1.02	4.1E-01
I-129	50	900 m ENE	0.60	2.4E-01
Pu-239	50	900 m ENE	69.3	2.8E+01
Am-241	50	900 m ENE	3130	1.3E+03
H-3	70	700 m E	14.3	5.7E+00
Kr-85	70	700 m E	0.53	2.1E-01
I-129	70	700 m E	0.31	1.2E-01
Pu-239	70	700 m E	36.4	1.5E+01
Am-241	70	700 m E	1640	6.6E+02
H-3	100	900 m E	7.21	2.9E+00
Kr-85	100	900 m E	0.27	1.1E-01
I-129	100	900 m E	0.16	6.0E-02
Pu-239	100	900 m E	18.4	7.4E+00
Am-241	100	900 m E	833	3.3E+02
H-3	150	1200 m E	3.14	1.3E+00
Kr-85	150	1200 m E	0.12	5.0E-02
I-129	150	1200 m E	0.06	3.0E-02
Pu-239	150	1200 m E	8.09	3.2E+00
Am-241	150	100 m NNE	222	8.9E+01

Table A2: Impact of stack height on dose location and required DF for a facility located in Tennessee.

Isotope	Stack Height (m)	Location of MEI	Effective Dose Equivalent (mrem)	DF (10%)
H-3	30	500 m NE	43.9	1.8E+01
Kr-85	30	500 m NE	3.18	1.3E+00
I-129	30	500 m NE	1.89	7.6E-01
Pu-239	30	500 m NE	221	8.8E+01
Am-241	30	500 m NE	10000	4.0E+03
H-3	50	700 m NE	17.2	6.9E+00
Kr-85	50	700 m NE	1.25	5.0E-01
I-129	50	700 m NE	0.74	3.0E-01
Pu-239	50	500 m S	88.5	3.5E+01
Am-241	50	500 m S	4030	1.6E+03
H-3	70	600 m S	9.54	3.8E+00
Kr-85	70	600 m S	0.69	2.8E-01
I-129	70	600 m S	0.41	1.6E-01
Pu-239	70	600 m S	51.8	2.1E+01
Am-241	70	100 m S	1450	5.8E+02
H-3	100	800 m S	5.17	2.1E+00
Kr-85	100	800 m S	0.37	1.5E-01
I-129	100	800 m S	0.22	9.0E-02
Pu-239	100	100 m S	28.4	1.1E+01
Am-241	100	100 m S	1450	5.8E+02
H-3	150	1000 m S	2.45	9.8E-01
Kr-85	150	1000 m S	0.18	7.0E-02
I-129	150	1000 m S	0.11	4.0E-02
Pu-239	150	100 m S	28.4	1.1E+01
Am-241	150	100 m S	1450	5.8E+02