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Simplified Improved Repository Risk Assessment Measure

Phase 2 Report

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ABSTRACT

The operation of nuclear reactors generates irradiated materials which must be disposed of in a mined repository or other deep geologic facility. The formal performance assessment of the numerous forms of waste that may be destined for these facilities can be resource intensive, so surrogate measures are often used for initial scoping calculations, such as waste volume, radioactivity, or radiotoxicity. Current surrogate measures are imperfect at capturing the risk associated with various radionuclides due to, in part, the mobility those materials have through the earth.

This study was conducted to progress work on alternative measures to capture the more significant risk drivers of various materials present, and to produce a Simple Improved Repository Risk Assessment Measure (SIRRAM).

Keywords

Disposal
Nuclear waste
Radionuclide
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1 INTRODUCTION

1.1 Summary of Previous Work

Estimates of risks from waste disposal in a geologic repository can be used for evaluating preferences for nuclear technology options; such evaluations typically involve characterizing and weighting multiple criteria. These risks can take many forms, such as the decay heat of different materials, the half-lives they will emit during, and the ease of transport through groundwater pathways. Preparing a repository performance assessment (PA) for multiple technology options can be impractical because of varying waste forms, differing source terms, and the absence of a specific repository site. Thus, the repository risk is typically addressed by surrogate measures that are simple to calculate, such as: waste volume, radioactivity, or radiotoxicity. Such surrogate measures, taken in isolation, are imperfect measures of repository risk. To address the need for a repository risk assessment measure that is accurate enough to be used during Research, Development, and Demonstration (RD&D), but simple enough to be calculated using the incomplete information typically available during RD&D, a preliminary study was undertaken to develop a Simple Improved Repository Risk Assessment Measure (SIRRAM)[1]. The results of the preliminary study were documented in a paper [Croff 2015] and are briefly summarized here.

The approach used in the preliminary study was to conduct an initial survey of performance assessments for planned repositories in a range of geologies to determine which radionuclides dominated the repository risk. The result was a fortuitous commonality among performance assessments: the list of radionuclides that dominate calculated risk is short and mostly contains the same radionuclides irrespective of the site or geology being assessed. To illustrate this point, results from 15 repository performance assessments for a variety of geologic media (granite, clay, salt, deep boreholes) were analyzed to identify the radionuclides that dominate risk at the time of peak exposure. Results of the analysis are given in [Croff 2015], which identifies the radionuclides that contribute at least 0.0001% of the sum of the peak dose rates, out to a 1-million-year decay time, to a member of the public from the normal evolution of an undisturbed repository containing spent LWR fuel.

The observations based on this analysis and the performance assessments underlying it are as follows:

- I-129 dominates long-term repository dose by at least a factor of five in all but one of the 15 studies examined and even there it remained a significant contributor to risk.
- The calculated repository risk of technologies producing the same amount of energy will differ according to the fission product yields for a particular neutron energy spectrum.

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[1] This report is focused on wastes such as spent nuclear fuel, high-level waste, and some greater-than-Class C (GTCC) wastes that are currently destined for a deep geologic repository. However, the concepts developed in this report should be applicable to other types of disposal technologies such as shallow-land burial but the importance of various radionuclides would require re-evaluation.

- C-14 and Cl-36 are produced by neutron captures in stable nitrogen and chlorine precursors, respectively. These precursors may be more common in the materials being considered for advanced reactor designs.

1.2 Description of the Phase 2 Follow-on Effort

This follow-on study builds on the results of the preliminary study to develop an improved measure of long-term repository risk for use in comparing nuclear repository technologies that will be simple to evaluate.

The preliminary study identified seven radionuclides that are relevant to repository risk. At the end of the preliminary study a number of issues remained to be addressed. This follow-on study addresses these issues with enhancements in several areas as follows:

1. *Obtaining available data on the concentration of activation product precursors in advanced nuclear systems*. The production rate of two radionuclides of interest (C-14 and Cl-36) depends on the concentration of their stable precursor radionuclides which, in turn, depends on the material being irradiated and how it is manufactured. Section 2 identifies a range of advanced reactor systems, the main materials (e.g., cladding, fuel) being irradiated, and typical concentrations of the precursor radionuclides.
2. *Determining the relative concentrations of significant fission product contributors from a number of actinides.* The production rate of the fission products of interest depend on fission yields and, in some cases, on the relevant cross sections which depend on the fissioning species and the reactor neutron spectrum. A comparison of the production rate as a function of fissioning species and neutron spectrum is provided in Section 3 as a basis for determining the implications of these parameters to the development of a SIRRAM.
3. *Update and analysis of the set of repository performance assessments underpinning a SIRRAM.* Fifteen PAs in seven source documents were used in the Phase 1 study. The set of PAs used in the preliminary study were supplemented by additional PAs which were analyzed to identify any additional important radionuclides (IRNs) and to qualitatively assess the cause(s) of variation in the order of radionuclide importance to calculated doses. These results are described in Section 4.
4. *Investigating how to determine weighting factors for each radionuclide considered in the SIRRAM.* The preliminary study used the peak future dose to a receptor (member of the public) as the measure of risk. However, the decay profile (e.g., the time an IRN reaches the receptor, the magnitude of the peak dose) of radionuclides vary depending on half-lives and its transport properties (e.g., solubility, sorption) from the source to the receptor. The results of investigating methods for accounting for these differences are discussed in Section 6 leading to weighting factors to allow a single value for the SIRRAM to be calculated.

2 MATERIALS AND ACTIVATION PRODUCT PRECURSORS IN ADVANCED NUCLEAR REACTOR SYSTEMS

In the preliminary study, the list of key radionuclides important to repository risk was derived from the disposal of LWR spent nuclear fuel (SNF). However, with the advent of interest in several advanced reactor technologies, novel materials will be utilized during reactor operations that will deviate from traditional LWRs. As a result, the SNF/HLW from advanced reactors placed in a repository has the potential to contain radionuclides that have not previously been accounted for in the initial assessment. The materials that will be of interest in the development of the improved SIRRAM will mainly be materials that will be exposed to neutron irradiation in advanced reactor systems (e.g., fuel elements, cladding, and coolant media). These novel materials have the potential to be precursors for activation products which are unique to advanced reactors and therefore potentially risk-significant in a repository setting. The following section outlines several advanced reactor concepts currently under development and their respective materials that may be potential precursors of radionuclides important to repository risk.

2.1 Advanced Reactors Being Considered

When evaluating which advanced reactor technologies to consider in developing the improved SIRRAM, the emphasis was placed on designs with significant differences with respect to LWRs. That is, designs that feature novel fuel types, cladding, or coolant media were the focus of the technologies being considered. Six advanced reactor designs that feature a variety of fuel elements, coolant media, and neutron spectra that will be considered in this study are as follows:

- Gas-Cooled Fast Reactor (GFR)
- Lead-Cooled Fast Reactor (LFR)
- Molten Salt Reactor (MSR)
- Sodium-Cooled Fast Reactor (SFR)
- Supercritical Water-Cooled Reactor (SCWR)
- High-temperature Gas-cooled Reactor (HTGR)

Of these reactor designs, five were considered for the improved SIRRAM, the exception being the SCWR concept. This is based on the assumption that the SCWR will feature light water coolant with either UO_2- or MOX-based fuels [Schulenberg 2014]. Because these are design features that are also common to LWRs, SWCRs were not considered further in the follow-on study, as the repository risk is likely to be similar to that presented by LWRs. The remaining Gen IV reactors are anticipated to utilize combinations of novel fuels/coolants/cladding materials and will be discussed further. In addition to advanced reactor systems, accident tolerant fuels (ATFs) and novel cladding materials for currently operating LWRs will also utilize materials that have not previously been utilized in LWR operations [Sena 2020]. These ATFs and novel cladding materials may also have the potential to be precursors for radionuclides important to repository risk, and they are discussed further in following sections.

2.2 Novel Materials Present in Advanced Reactor Systems

From the advanced reactor technologies identified in Section 2.1, a literature survey was conducted to identify novel materials potentially present within the respective designs. Table 1 summarizes the results of the literature survey, and the following subsections provide descriptions of each of the advanced reactor technologies and their novel materials.

Table 1. Novel materials anticipated for advanced reactor technologies

Technology	Reference Design	Fuel Type	Other Novel Material(s)	Reference(s)
Gas-Cooled Fast Reactor	Energy Multiplier Module (EM^2), General Atomics	UC_{1+x} (LEU/DU)	SiC cladding	Choi 2020
	French Alternative Energies and Atomic Energy Commission (CEA) GFR fuel design R&D	U-Pu-C	Mixed SiC/metal cladding • Ta metal alloy liner • Niobium metal alloy liner	Zabiégo 2013
Lead-Cooled Fast Reactor	BREST	(U,Pu)N	Lead Coolant	Khalil 2000
	INL/MIT actinide burner	U-Th-Pu-MA[a] Nitride or U-Th-Pu-MA-Zr metal	Lead-Bismuth Eutectic	Buongiorno 2001
	European Lead System (ELSY)	$(Pu,U)O_2$ MOX	Lead Coolant	Alemberti 2011
Molten Salt Reactor	Fluoride Salt-Cooled High Temperature (FHR), Kairos Power (fluoride-cooled)	U-C-O (TRISO[b])	$2LiF:BeF_2$ (Flibe) coolant	Kairos Power LLC, 2018
	Generic MSR Structural Material Study	Fluoride-based[c]	NiMo-Y_2O_3 alloys for structural materials	Li 2022

Table 1 (continued). Novel materials anticipated for advanced reactor technologies

Technology	Reference Design	Fuel Type	Other Novel Material(s)	Reference(s)
	Molten Salt Reactor Experiment (MSRE) (fluoride-fueled)	$LiF-BeF_2-ZrF_4-UF_4$	$2LiF:BeF_2$ (Flibe) coolant	Haubenreich 1970
	Generic MCFR Design Study (chloride-fueled)	$NaCl-UCl_3$	-	Mausolff 2021
	BN-600	UO_2	Liquid sodium coolant	Aoto 2014
Sodium-Cooled Fast Reactor	AMORUUUC	$(Pu,U)O_2$ MOX	Liquid sodium coolant	Croff 1981
High-temperature Gas-cooled Reactor	X-Energy	U-C-O (TRISO)	In-core ceramic composites (C/C; SiC/C;SiC/SiC)	Mulder 2021; IAEA, 2010
Advanced LWR	Framatome	Chromia-doped UO_2	Chrome-coated cladding	Delafoy 2018
	Global Nuclear Fuel	UO_2	Fe-Cr-Al cladding, ARMOR coating	Lin 2018
	INL	U_3Si_2	-	Wagner 2019
	General Atomics	UO_2	SiC cladding	Deck 2019

[a] MA = minor actinides

[b] Electric Power Research Institute. (2019). Uranium Oxycarbide (UCO) Tristructural Isotropic (TRISO) Coated Particle fuel Performance. Report No. 3002015750.

[c] Alloy samples corroded in FLiNaK salt

2.2.1 Gas-Cooled Fast Reactors (GFRs)

For GFR systems, the Energy Multiplier Module (EM^2) from General Atomics is a design that is under active development [Choi 2020]. The design features uranium carbide (UC) fuel pellets which are comprised of low-enriched uranium (LEU) and depleted uranium (DU) carbide. The cladding for the fuel pellets consists of a silicon carbide (SiC) composite that encases the pellets. It features a multi-layer SiC composite that contains SiC fibers in the primary layer and a monolith SiC layer for the outer portion. A slightly different fuel element design was proposed by the French Alternative Energies and Atomic Energy Commission (CEA) wherein the fuel is comprised of a mixed uranium-plutonium carbide, and the SiC cladding is bolstered with tantalum and/or niobium alloys in a ‘metallic skeleton’ arrangement [Zabiego 2013].

2.2.2 Lead-Cooled Fast Reactors (LFRs)

The BREST LFR concept developed by the Russian Federation utilizes nitride-based fuel pellets encapsulated in stainless steel cladding [Khalil 2000]. The design feature that distinguishes this concept, however, is the usage of a pure liquid lead coolant. The European Lead Fast Reactor (ELSY) also features pure lead coolant but includes MOX-based fuel [Alemberti 2011]. Lead-bismuth eutectic coolants have also been proposed in designs such as the INL/MIT actinide burner [Buongiorno 2001]. These coolants have the potential to be precursors for activation products that have not previously been generated in LWRs. Additionally, the nitrides featured within some of the fuel element designs have the potential to generate activation products unique to LFRs.

2.2.3 Molten Salt Reactors (MSRs)

MSR systems feature a variety of designs that will feature combinations of fuels and/or coolant media that will significantly deviate from materials in LWR operations. Amongst MSRs, there are three primary designs that are being considered: solid-fueled with fluoride-salt coolant, liquid-fueled with fluoride-salt, and liquid-fueled with chloride-salt. An example of the solid-fueled, fluoride-coolant variation is Kairos Power's Fluoride Salt-Cooled High Temperature Reactor (KP-FHR) [Kairos Power LLC, 2018]. The KP-FHR features 'pebble' type tri-structural isotropic (TRISO) fuel elements which are comprised of high-assay low enriched uranium kernels in the form of U-C-O. The fuel kernel is surrounded by several layers comprised of different materials including graphite, pyrolytic carbon, and silicon carbide. In addition to the TRISO fuel, the KP-FHR will also utilize a FLiBe salt ($2LiF{:}BeF_2$) mixture for the coolant medium. Any nitrogen dissolved in the FLiBe salt will produce ^{14}C [Ying 2019] which has been identified as an IRN.

For both the fluoride- and chloride-fueled MSR design variations, activation and fission products will be generated continuously within the fuel-salt matrix. The sole example of operational MSR experience was with the Molten Salt Reactor Experiment (MSRE), which featured the $LiF\text{-}BeF_2\text{-}ZrF_4\text{-}UF_4$ fuel and FLiBe coolant [Haubenreich 1970]. For chloride-based designs, several variations of the fuel salt exist, but the $NaCl\text{-}UCl_3$ matrix seems to represent the fuel salt system that will most likely be utilized for commercial applications [Mausolff et al., 2021]. Because of this, the $NaCl\text{-}UCl_3$ fuel salt was chosen to be representative of chloride-fueled MSRs. With any chloride-based design, however, activation of ^{35}Cl to the long-lived ^{36}Cl isotope ($t_{1/2} = 3.01 \times 10^5 years$) will occur unless the chloride in the fuel salt is enriched to isotopically pure ^{37}Cl [Riley 2018].

Apart from the salts, MSRs may also feature novel materials in the structural components to ensure high tolerance in corrosive and high temperature environments. For example, a recent study by [Li 2022] has shown that Y_2O_3 dispersion-strengthened NiMo-based alloys show improved swelling and corrosion resistance when compared to the Hastelloy N alloy used in the MSRE. As developments continue to be made with respect to enhanced structural materials, such as the ones just mentioned, activation products could be introduced and impact the radionuclides considered for evaluating repository risk.

2.2.4 Sodium-Cooled Fast Reactors (SFRs)

SFRs represent perhaps the most mature advanced reactor design, with designs such as the currently operational Russian BN-600 and BN-800, and previously operational systems such as the Prototype Fast Reactor (PFR) in the United Kingdom and the Phénix and Superphénix reactors in France [Aoto 2014]. Because of its current operational status, the BN-600 design was chosen as a reference SFR in Table 1. The fuel element materials are comprised of UO_2, and liquid sodium is used as the coolant. The liquid sodium coolant represents the material that is of potential interest for determining activation products not addressed in prior studies. The second SFR design referenced in Table 1, the AMORUUUC concept, was described in [Croff 1981] to model liquid-metal fast breeder reactors in the ORIGEN2 point depletion computer program. The AMORUUUC model utilizes a U-Pu oxide fuel, and a representative neutron flux for a fast reactor was determined using a depletion calculation from this model (see Section 3.2.1).

2.2.5 High-Temperature Gas-Cooled Reactors (HTGRs)

The high temperature gas-cooled reactor (HTGR) includes several designs that are used for both commercial and experimental applications [IAEA 2010]. They include enhanced safety functionality and high operating temperatures. Several HTGR systems are currently being designed; the X-Energy Xe-100 concept was chosen as a representative design for the HTGR due to its design maturity [Mulder 2021]. The Xe-100 features graphite fuel elements containing TRISO fuel with a helium coolant. Other novel materials that are likely to be included within this design (and for other HTGR designs in general) are in-core ceramic composites made of SiC and C-based materials that can withstand high temperatures and irradiation damage [IAEA 2010].

2.2.6 Advanced LWRs

For currently operating LWRs, there are several novel materials that could be used in accident-tolerant fuels (ATFs) and improved claddings. A summary of expected milestones for these applications in the near-term is given by [Sena 2020]. Some examples of anticipated novel materials for LWRs are as follows:

- Framatome’s Chromia-doped UO_2 fuel + Chrome-coated cladding [Delafoy 2018]
- Global Nuclear Fuel’s Fe-Cr-Al cladding + ARMOR coating [Lin 2018]
- INL’s U_3Si_2 fuel element [Wagner 2019]
- General Atomics’ SiC Cladding [Deck 2019]

2.3 Consideration of Radionuclides Present in Advanced Systems

Given the novel materials identified in the various advanced reactor technologies in the previous section, several activation product precursor materials can be identified that may contribute to repository risk. From the information gathered in Table 1, the elements that may be more prevalent in advanced nuclear systems, when compared to traditional LWRs, are summarized as:

- Gas-Cooled Fast Reactor
  - Carbon and silicon from fuel cladding and assembly structural materials
  - Tantalum and/or niobium from fuel cladding and assembly structural materials
- Lead-Cooled Fast Reactor
  - Lead/lead-bismuth eutectic coolant
  - Nitrogen from nitride-based fuel elements
- Molten Salt Reactor
  - Carbon from TRISO fuel elements (for FHR-type MSRs)
  - Fluoride, lithium, and beryllium from FLiBe coolant/fuel salt
  - Chlorine from chloride-based fuel salt
  - Molybdenum from structural materials
- Sodium-Cooled Fast Reactor
  - Sodium from coolant
- High-temperature Gas-cooled Reactor
  - Carbon from TRISO fuel elements
  - Carbon and silicon from fuel cladding and structural materials
- Advanced LWR
  - Chromium from certain ATFs and cladding
  - Silicon from certain ATFs
  - Carbon and silicon from cladding

From this list, it is apparent that a number of elements will be present within advanced reactor systems that are not present in LWRs. In particular, carbon, silicon, lead, bismuth, fluorine, lithium, beryllium, molybdenum, chlorine, and sodium are either anticipated across several designs (e.g., carbon and silicon) or are going to be present in much larger quantities than in LWRs (e.g., chlorine in a chloride-based MSR or sodium in an SFR). From these elements, potential activation products can be identified that are generated after neutron capture of the precursor material. A list of common radionuclides with significant radioactive inventories present in six performance assessments of waste disposal sites is given in [Baudoin 2000]. Using this list, and the list of elements anticipated to be prevalent in advanced nuclear systems, the potentially important activation products resulting from the listed elements can be identified (shown below in Table 2).

Table 2. Potentially important activation products from advanced nuclear systems

Activation Product Precursor Identified in an Advanced System	Corresponding Activation Product Listed in [Baudoin 2000]	Half Life
Nitrogen	C-14	5700 years
Silicon	Not listed	-
Lead	Pb-210	22 years
Bismuth	Not listed[a]	-
Fluorine	Not listed	-
Lithium	H-3	12 years
Beryllium	Be-10	1.4×10^6 years
Molybdenum	Mo-93	4000 years
Chlorine	Cl-36	3×10^5 years
Sodium	Not listed	-

[a] The primary activation product of concern with the lead-bismuth eutectic is Po-210 [Buongiorno 2001], which is relatively short-lived ($t_{1/2}$ = 140 days) and, thus, not a common radionuclide of concern in a repository setting

Of the activation products listed in Table 2, C-14 and Cl-36 were previously identified as IRNs in the preliminary study of the SIRRAM. Since these radionuclides have already been identified as being significant to risk in a repository, and since they are anticipated to also be prevalent for advanced nuclear systems, they will be further considered in the development of the improved SIRRAM. Several additional radionuclides that were not considered previously, however, have been identified:

- Pb-210
- H-3
- Be-10
- Mo-93

Of the newly identified activation products, each has a sufficiently long-lived half-life to warrant consideration with the exception of H-3 (tritium). Tritium production is enabled by lithium precursors, and it is produced by the reaction $Li^6(n,\alpha)H^3$. This is anticipated to be most prevalent in fluoride-based MSR systems where FLiBe is used as the primary fuel and/or coolant salt. Despite its prevalence in certain MSR systems, however, tritium is not considered further in the development of the improved SIRRAM due to its short half-life (12 years) in the context of repository risk. When evaluating repositories in the long-term, the main risk-contributing radionuclides have half-lives which are orders of magnitude greater than tritium; that is, tritium will decay to negligible amounts after only 100 years (which is equivalent to ~8 half-lives). To determine if the remaining radionuclides might significantly contribute to repository risk, they are assessed in terms of dose to a potential off-site receptor.

In a performance assessment conducted on the Yucca Mountain geologic repository, several of the radionuclides previously identified were also listed [Mohanty 2002]. Mo-93 and Pb-210 were radionuclides that were common to both lists, and of these, only Pb-210 contributed to a cumulative dose greater than 10^{-8} mSv/year. The dose to an off-site receptor was calculated to 100,000 years, and Mo-93 did not significantly contribute to the overall dose. Additionally, a set of screening criteria was developed by [SNL 2007] in which the radionuclides important to the performance of the Yucca Mountain Project were assessed and analyzed for time periods ranging up to 1 million years and for four release scenarios to off-site receptors. Of the four scenarios analyzed, three pertained to groundwater releases. Of the screened radionuclides that were deemed significant to the performance of the repository, molybdenum was not included because its relative importance in terms of dose to a receptor, which was determined by taking the product of the activity inventory and several screening factors, did not contribute more than 5% of the total of the screening products within a set of screened radionuclides. Based on the results from these two studies, Mo-93 was not considered further as a radionuclide that would significantly contribute to repository risk.

The Be-10 radionuclide is not commonly seen in repository performance assessments, but it is listed on one occasion as being potentially important in [Baudoin 2000], and it was shown in two PAs (see Table 11 Items #21 and #28) for a repository in clay. The source of the Be-10 is from irradiation of startup neutron sources—some of which are composed of a relatively-short lived alpha-emitting actinide (e.g., Po-210, Ra-226, Pu-238)—and beryllium that produce neutrons via an (alpha, n) reaction. The result of the ANDRA PA [ANDRA 2005] was that Be-10 did not appear as a dose contributor in the reference cases because it was at least 4 orders of magnitude less than the largest contributor and fell below the lowest dose rate on the ordinate (10^{-10} Sv/year). In some sensitivity analyses Be-10 was calculated to have a peak dose about 0.001% of the maximum for the SNF. In [Chornoboy 2018] Be-10 was calculated to contribute about 0.01% of the total peak dose. On this basis, Be-10 is not included as an IRN in this analysis.

2.4 Activation Product Precursor Concentrations

To determine the overall impact to repository risk, it is also necessary to have estimates of the concentrations of each of the activation product precursors in each of the advanced nuclear systems previously identified. However, accurate estimations of the precursor concentrations are not possible since many of the designs and material compositions are currently in early stages of development or are proprietary. Therefore, measured values of activation product precursors found during the development of the ORIGEN2 code have been adopted and used them in the resulting models. The values adopted for the present study are shown in Table 3.

Table 3. Activation product precursor concentrations[a]

Element	Atomic Number	Concentration (g/MTHM)[b]	Element	Atomic Number	Concentration (g/MTHM)[b]
Lithium	3	1.0	Manganese	25	1.7
Boron	5	1.0	Iron	26	18.0
Carbon	6	89.4	Cobalt	27	1.0
Nitrogen	7	25.0	Nickel	28	24.0
Oxygen	8	134, 454[c]	Copper	29	1.0
Fluorine	9	10.7	Zinc	30	40.3
Sodium	11	15.0	Molybdenum	42	10.0
Magnesium	12	2.0	Silver	47	0.1
Aluminum	13	16.7	Cadmium	48	25.0
Silicon	14	12.1	Indium	49	2.0
Phosphorus	15	35.0	Tin	50	4.0
Chlorine	17	5.3	Gadolinium	64	2.5
Calcium	20	2.0	Tungsten	74	2.0
Titanium	22	1.0	Lead	82	1.0
Vanadium	23	1.0	Bismuth	83	0.4
Chromium	24	4.0			

[a] This table was reproduced from [Croff 1978].

[b] Parts of element per million parts of initial heavy metal by weight.

[c] Stoichiometric quantity for (Pu,U)O_2 fuel; use 137, 931 grams per metric ton of heavy metal (g/MTHM) for thorium-based fuels.

3 RELATIVE PRODUCTION RATES OF IMPORTANT CONTRIBUTORS TO REPOSITORY RISK IN THERMAL AND FAST NEUTRON SPECTRA FROM MAJOR ACTINIDES

This section analyzes the relative production rate of radionuclides that are dominant, frequent, or sometimes relevant to repository risk. The reason for conducting such an analysis is to provide the basis for comparing the production rate of the IRNs from various fissionable materials. This comparison provides potentially useful insights for prioritizing development of advanced reactor concepts, and the extent to which differences in the concentration of the important contributors in the repository waste need to be accounted for when developing a SIRRAM.

The analysis begins by identifying the important radionuclides to be analyzed and the key parameters that could control their production rate which is done qualitatively in Section 3.1. The analysis approach, assumptions, and parameters needed by the ORIGEN2 computer code [Croff 1980a], which is used to calculate the radionuclide concentrations, are then established in Section 3.2. The results are then analyzed in Section 3.3 leading to an intermediate set of conclusions and recommendations in Section 3.4.

3.1 Identify the Variables to be Analyzed

The fissionable materials considered are the actinides U-233, 235, and 238, Pu-239, 241, and Th-232. The fission and activation product IRNs considered in this analysis began with a list developed in the preliminary study described in Section 1.1 and considered the addition of radionuclides as described in Section 2. The IRN production rates considered in this analysis are calculated using cross sections for a thermal neutron spectrum from a pressurized-water reactor (PWR) and for a fast neutron spectrum from a sodium-cooled fast reactor (SFR) with the actinides being in oxide form in both cases. The list of IRNs, their half-lives, and their capture cross sections are given in Table 4.

Table 4. Radionuclides Important to repository risk, their half-lives, and cross-sections

Important Radionuclide	Half-life, years[a]	Thermal Capture cross section, barns[b]	Fast Capture Cross Section, barns[c]	Precursor Isotope (FI = Fissionable Isotope)	Precursor Thermal Capture cross section, barns[b]	Precursor Fast Capture Cross Section, barns[c]
C-14	5700	8.54E-08	6.94E-10	N-14	1.81E-01	1.39E-02
Si-32[d]	153	1.23E-03	3.33E-04	Si-30 Si-31	1.26E-02 4.15E-02	1.64E-04 3.33E-04
Cl-36	301,000	8.54E-01	6.94E-03	Cl-35	3.897	2.62E-02
Se-79	326,000	3.37E-01	2.47E-03	FI	Varies	Varies
Tc-99	211,100	9.32E+00	4.52E-01	FI	Varies	Varies
Sn-126	230,000	3.08E-02	5.48E-02	FI	Varies	Varies
I-129	15,700,00	5.02E+00	3.93E-01	FI	Varies	Varies
Cs-135	2,300,000	2.48E+00	6.56E-02	FI	Varies	Varies

[a] Source: [NNDC 2022]

[b] Cross section and flux sources: [Croff 1978; Croff 1983].

[c] Source: [Croff 1983]

[d] Source: Inferred from comparison of cross-section plots for Si-31 and Si-32 as a function of energy in [Kopecky 1997], the thermal cross-section of Si-32 is 3% of the Si-31 cross section and the fast Si-32 cross section is equal to that of Si-31. Uncertainty is large.

One factor that requires early adoption is the measure of IRN production rate to result in a defensible comparison. Using traditional measures, such as mass or volume concentration (e.g., grams per MTHM or liter), is not appropriate because the IRN concentration would vary with both burnup and fissionable material mass concentrations, which are dependent on design assumptions that are unknown for most advanced reactors and are subject to revision during development. The same is true of other potential measures such as radioactivity, because they are simply the mass or volume concentration multiplied by a constant conversion factor.

A defensible measure needs to reflect the IRN production rate that is characteristic of each particular fissionable isotope and the neutron spectrum that is consistent with the particular advanced reactor concept, but not be sensitive to evolving or unknown design assumptions. The high-level objective of the SIRRAM can be viewed as informing reactor development and design decisions by providing insights that lead to minimizing the amount of each IRN produced per unit of energy produced. Thus, the mass (grams) of each IRN resulting from the production of one thermal megawatt (MW_t) of fission energy produced is the first component of the measure adopted in this study. It is readily calculated by dividing the mass concentration of an IRN after some irradiation duration by the burnup of the fuel, as calculated by the ORIGEN2 model at that time, with the resulting unit being g/MW_td.

However, as discussed above, the amount of each IRN produced is also proportional to the amount of some precursor species. In the case of activation products the precursor is one or more stable isotopes (e.g., Cl-35 in the case of the Cl-36 IRN). For fission products, the precursor is one or more of the fissionable isotopes identified in Section 3.1, the concentration of which, when multiplied by a fission cross section, a fission product yield, a flux, and conversion constants, results in the IRN production rate. Again, the IRN production rate is proportional to the amount of the actinide isotope producing energy. In both cases, the amount of precursor being irradiated is taken into account by dividing the IRN production rate having units of g/MW_td by the amount of precursor isotope being irradiated to normalize the production rate to a mass of one part per million of the precursor[2] with the resulting measure having units of g/(MW_td-ppm).

3.2 Establish the Analysis Approach and Assumptions

The most obvious and useful comparison of the IRN production rate from each fissionable isotope would be to simply to calculate the mass of each IRN produced by irradiating fissionable isotopes using a reactor physics code that can simulate radionuclide buildup and depletion. Doing so raises a number of issues:

1. What depletion code to use.
2. How to address the fact that real fuels are a mixture of fissionable materials, which means that the IRN production rate cannot be separately associated with each fissionable material.
3. How to obtain an appropriate neutron flux value for each reactor and fuel type.
4. Avoiding the buildup of additional fissionable materials during irradiation.
5. Depletion of precursor isotopes.

These issues are addressed in the following sub-sections.

3.2.1 Depletion Code Selection

A rigorous radionuclide buildup and depletion approach would involve use of reactor physics code systems such as SCALE to model the fuel lattice or equivalent in a particular reactor design to calculate relevant cross sections and fluxes and then calculate radionuclide buildup and depletion in various fissionable materials. This is not possible in this analysis because the reactor designs are typically in early stages and/or proprietary. Also, the detailed results from the reactor code systems (and the associated labor and computational resources) are unnecessary for the purposes at hand: developing a SIRRAM and informing early-stage R&D decisions.

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[2] In this study the precursor is defined to be the natural element containing the precursor isotope of interest. Some reactors may involve enriched materials (e.g, chlorine enriched in Cl-37) so the precursor material concentration or composition would need to be adjusted accordingly.

Instead, the ORIGEN2 code [Croff 1980a] is used to calculate radionuclide buildup and depletion for representative thermal and fast reactors. The thermal reactor cross sections are from a model of a PWR using oxide fuel [Croff 1978; Croff 1980b] and the fast reactor cross sections are from an advanced sodium-cooled fast reactor with oxide fuel [Croff 1983], as discussed above in Section 3.1.

3.2.2 Disentangling Important Radionuclide Production

The overall approach to comparing the production rate of the IRNs listed in Table 4 is to irradiate one metric ton initial heavy metal (MTIHM) of pure fissionable material oxides listed in Section 3.1 using ORIGEN2 and cross sections appropriate for a thermal or fast neutron spectrum with a particular fuel type (uranium- or thorium-based). A straightforward approach would involve taking representative fissionable material mixtures and specific power levels in representative reactors (such as those in the ORIGEN2 models adopted in the previous section), calculating the resulting concentration of each IRN, and analyzing the results to obtain insights on their production rate. Attempting to do so leads to a challenge: the fissionable isotope in a nuclear fuel is seldom present as a single isotope. Moreover, the initial fuel is composed of multiple fissionable isotopes. For example, the initial fissionable isotopes in fresh LWR fuel are typically U-235 and U-238, and later in life Pu-239 and 241 make significant contributions. As a consequence, each IRN production rate is a complex function of the differing fission product yields and fission cross sections of each fissionable isotope, the changing fissionable isotope concentrations, and the initial fissionable isotope concentration. The source of each IRN is entangled because the Bateman equations solved by ORIGEN2 yield just a single IRN concentration at the end of each irradiation step, and the fraction of the production rate cannot be separately attributed to each fissionable isotope.

The first approach adopted for the comparison of fissionable materials is to irradiate each fissionable isotope separately which then results in the mass concentration of each IRN that is solely attributable to that isotope. The mass concentration can readily be converted to the adopted measure of IRN production rate [g/(MW$_t$-ppm)] using the burnup calculated by ORIGEN2 for the underlying reactor models (see last paragraph of Section 3.2.1 and the initial amount of the IRN precursor). The amount of each fissionable material precursor is assumed to be one MTIHM, and the activation product precursors were assumed to have the isotopic abundance of the naturally occurring element in concentrations taken from [Croff 1978] (see Section 2.4).

3.2.3 Avoid the Potential for Widely Varying Neutron Fluxes

Separately irradiating each pure fissionable isotope oxide, as assumed in Section 3.2.2, leads to another challenge. Typically, irradiation of fissionable material in ORIGEN2 is based on an assumed specific power (MWt/MTHM) for a mixture of actinide isotopes and a sufficient irradiation duration to achieve the desired (or allowable) burnup – with the neutron flux being a calculated result. However, if the particular specific powers from the ORIGEN2 reactor models are assumed and applied to the various individual fissionable isotopes, the result would be very different neutron fluxes for each isotope, with higher fission cross sections leading to lower neutron fluxes. For example, the U-235 fission cross section in a PWR fueled with LEU oxide is about 500 times larger than the U-238 fission cross section. As a result, the flux in the U-238 irradiation would be about 500 times that for the U-235 irradiation to allow the U-238 to achieve the same burnup as the U-235 in the same duration. In reality, neither flux would be representative of the mixture, and the actual flux associated with the reactor model would be an average of the two fluxes weighted by the concentration of the precursor isotopes in the mixture.

The differing fluxes would render the calculated IRN production rates invalid for the purposes of comparison because: (1) the depletion of the fissionable isotopes by neutron absorption other than fission, (2) the depletion of the fission and activation products precursors, and (3) the production rate of the IRNs from precursors are all linearly dependent on the differing fluxes. Additionally, the resulting IRN production rates would be nearly the same because the yields are similar and the burnup is the same. This is not what occurs in reactor fuel composed of a mixture of fissionable materials where the burnup of each fissionable isotope differs because its fission cross section differs.

The solution adopted for the purpose of comparing the concentration of IRNs produced by each fissionable material is to use an alternative option available in ORIGEN2: specify the magnitude of the flux for each class of fuel and irradiate the fixed amount of each pure actinide precursor isotope using that flux. This approach has typically been used in ORIGEN2 calculations where the fuel matrix is irradiated by specifying an exact power and then the resulting fluxes are used to separately irradiate material containing little or no fissionable material (e.g., cladding). In doing so, the actinides are exposed to a flux that is representative of what would occur in a reactor containing a mixture of the fissionable materials. The fluxes assumed for the purpose of the comparison and their sources with associated fuel burnups are given in Table 5.

3.2.4 Avoiding Substantial Buildup of Fissionable Isotopes Other Than the Initial Isotope

Yet another challenge arises even if the irradiation calculation begins with a pure fissionable isotope. Irradiation of a fissionable isotope results in some of it being fissioned, but neutron capture also leads to the production of new fissionable isotopes. Fission of the new isotopes essentially re-creates the entanglement challenge described in Section 3.2.2, since different fissionable isotopes have different fission product yields. This challenge does not arise with the activation products, the precursors of which are stable isotopes. Overcoming this challenge involved a twofold approach. The first step was to assume a relatively short 20-day irradiation time to minimize the production of new fissionable materials. While some additional fissionable material having different fission product yields will still be produced, the amounts are small, so the IRN production rates are not significantly affected. The second step implemented is to set the neutron capture cross sections for the individual fissionable isotopes to zero by editing the ORIGEN2 cross section library. This technique leads to virtually no additional fissionable isotope production except for small amounts resulting from reactions such as (n,2n) and (n,3n).

Table 5. Assumed neutron fluxes for calculating important radionuclide production rates from four fuel types

Type of Reactor Fuel	Thermal Spectrum Flux: n/cm^2-sec Burnup: MWd/MTIHM	Fast Spectrum Flux: n/cm^2-sec Burnup: MWd/MTIHM
Fissile: U-235 Fertile: U-238	3.25E+14[a] 750 (20 days irradiation)	5.15E+15[c] 2465 (20 days irradiation)
Fissile: U-233 Fertile: Th-232	3.21E+14[b] 750 (20 days irradiation)	4.01E+15[d] 2706 (20 days irradiation)[3]
Fissile: Pu-239 Fertile: U-238	3.25E+14[a] 750 (20 days irradiation)	5.15E+15[c] 2465 (20 days irradiation)
Fissile: Pu-241 Fertile: U-238	3.25E+14[a] 750 (20 days irradiation)	5.15E+15[c] 2465 (20 days irradiation)

[a] Source: [Croff 1978]
[b] Source: [Croff 1980b]
[c] Source: [Croff 1981]
[d] Source: [Croff 1981]

3.2.5 Depletion of Fissionable Material

Yet another potential challenge is depletion of the initial fissionable isotope which results in a declining production rate of IRNs. The importance of this effect is somewhat mitigated by the two steps described in the previous section. For example, after the 20d irradiation in a thermal reactor, the highest fissionable isotope depletion was for Pu-241 where 91% of the initial Pu-241 remained.

3.2.6 Summary of ORIGEN2 Calculations

The buildup and depletion of radionuclides in one MTIHM of each of the six fissionable isotopes considered in this study were calculated in ORIGEN2 using the assumptions listed below.

- ORIGEN2 decay data were taken from a standard library that is independent of the type of nuclear material or the neutron spectrum [Croff 1979].
- Cross sections for the thermal spectrum calculations were based on a PWR reactor using LEU fuel for U-235, U-238, and bred plutonium isotopes [Croff 1978] and using thorium oxide fuel enriched by U-233 that had been denatured with U-238 [Croff 1980b].
- Cross sections for the fast spectrum calculations were based on a SFR using depleted uranium oxide fuel enriched with plutonium for U-235, U-238, and the plutonium isotopes (AMORUUUX core region); thorium oxide fuel was enriched with recycled U-233 (AMO0TTTX core region) [Croff 1981].
- The neutron fluxes given in Table 5 were used and were assumed to be the same for all actinides. The concentration of fission product precursors (fissionable elements) was assumed to be one MTIHM.

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[3] The values for U-233/Th-232 differ slightly because of differences in the fission cross sections.

- The irradiation duration was 20 days to minimize the production of new fissionable materials.
- Fissionable material neutron capture cross sections have been set to zero as described in Section 3.2.4 to make the differences in production rates among the fission product precursors more transparent.
- The ORIGEN2 results in terms of g/MTIHM of each fissionable isotope were mined to retrieve the mass concentrations of the IRNs.
- The mass concentrations were copied into a spreadsheet which was used to divide each mass concentration by the burnup shown in Table 5 and the assumed concentration of the precursor isotope to yield the normalized production rate of each IRN in terms of g/(MW_td-ppm). These results are discussed in the next section.

3.3 Analysis of Results

This section will undertake three types of comparisons:

1. The normalized production rate of IRNs from the six fissionable isotopes considered in this study separately in thermal and fast neutron spectra.
2. The normalized production rate of IRNs from the six fissionable isotopes considered in this study addressing differences between thermal and fast neutron spectra.
3. The production rate of the IRNs from some representative fuel compositions containing multiple fissionable isotopes in thermal and fast neutron spectra.

To reiterate, when considering the results presented throughout Section 3, the results given are production rates that do not account for retardation during transport through the geosphere and the relative toxicity of the various IRNs.

3.3.1 Normalized Production Rate of the IRNs

The normalized production rate of the IRNs from each of the six fissionable isotopes considered is summarized in Table 6. The grams of IRN produced per ppmw of natural precursor (C-14, Si-32, Cl-36) in Table 6 is helpful for comparison in non-standard fuel types (e.g. molten salt), while a later table will provide strictly g/MWd. These results are included because they are used later in Section 5.5.3 to estimate the IRN production rate from fuels composed of mixtures of fissionable isotopes.

A more transparent comparison of the relative IRN production rates was developed by calculating the average production rate of each fission product IRN from the four fissile isotopes and two fertile isotopes and then dividing this value by the IRN production rate for each fissile or fertile isotope and expressing the result as a percentage. The result is a measure of the extent to which each fissionable isotope produces a larger or smaller portion of each IRN, i.e., values less than 100% account for a comparatively smaller portion of the production rate and a value greater than 100% account for a comparatively larger portion of the production rates. These results are shown in Table 7.

Table 6. Production rate of radionuclides important to repository risk assessment

IRN	Precursor, ppm (FI = Fissionable Isotope)	Production rate, grams of IRN produced per ppmw of natural precursor element OR actinide isotope per MW$_t$d produced by fuel						Fissile IRN Production Rate		Fertile IRN Production Rate		Fert/ fiss Avg, %
		U-233	U-235	Pu-239	Pu-241	U-238	Th-232	Average	Std Dev ±%	Average	Std Dev ±%	
Thermal Neutron Spectrum												
C-14	N: 25.0	1.46E-07	1.64E-07	1.64E-07	1.64E-07	1.64E-07	1.46E-07	1.60E-07	5%	1.55E-07	6%	97.19%
Si-32	Si: 12.1	3.82E-15	4.18E-15	4.18E-15	4.18E-15	4.18E-15	3.82E-15	4.09E-15	4%	4.00E-15	4%	97.82%
Cl-36	Cl 5.3	1.98E-06	2.24E-06	2.24E-06	2.24E-06	2.24E-06	1.98E-06	2.17E-06	5%	2.11E-06	6%	96.99%
Se-79	FI 10^6	1.92E-08	6.01E-09	1.27E-08	5.86E-09	2.05E-11	5.28E-12	1.10E-08	50%	1.29E-11	59%	0.12%
Tc-99	FI 10^6	6.44E-07	6.84E-07	1.67E-06	1.75E-06	1.45E-09	1.83E-10	1.19E-06	44%	8.18E-10	78%	0.07%
Sn-126	FI 10^6	5.39E-08	1.31E-08	8.82E-08	4.28E-08	2.47E-10	5.27E-12	4.95E-08	54%	1.26E-10	96%	0.25%
I-129	FI 10^6	3.01E-07	1.15E-07	5.74E-07	3.43E-07	5.89E-10	3.59E-11	3.33E-07	49%	3.13E-10	89%	0.09%
Cs-135	FI 10^6	3.12E-07	2.61E-07	7.14E-07	7.11E-07	5.29E-10	1.27E-10	5.00E-07	43%	3.28E-10	61%	0.07%
Fast Neutron Spectrum												
C-14	N: 25.0	1.97E-07	2.55E-07	2.55E-07	2.55E-07	2.55E-07	1.97E-07	2.41E-07	10%	2.26E-07	13%	93.99%
Si-32	Si: 12.1	6.02E-16	1.15E-15	1.15E-15	1.15E-15	1.15E-15	6.02E-16	1.01E-15	23%	8.75E-16	31%	86.50%
Cl-36	Cl 5.3	4.26E-08	7.28E-08	7.28E-08	7.28E-08	7.28E-08	4.26E-08	6.52E-08	20%	5.77E-08	26%	88.41%
Se-79	FI 10^6	3.86E-09	1.49E-09	8.62E-10	1.27E-09	4.33E-11	8.69E-12	1.87E-09	63%	2.60E-11	67%	1.4%
Tc-99	FI 10^6	1.09E-07	1.29E-07	1.24E-07	1.67E-07	3.27E-09	3.02E-10	1.32E-07	16%	1.78E-09	83%	1.35%
Sn-126	FI 10^6	1.69E-08	6.82E-09	8.71E-09	9.67E-09	5.18E-10	8.65E-12	1.05E-08	36%	2.63E-10	97%	2.50%
I-129	FI 10^6	6.59E-08	3.41E-08	4.26E-08	5.08E-08	1.29E-09	5.91E-11	4.83E-08	24%	6.74E-10	91%	1.40%
Cs-135	FI 10^6	2.24E-07	2.31E-07	2.53E-07	3.16E-07	5.54E-09	9.13E-10	2.56E-07	14%	3.23E-09	72%	1.26%

Table 7. Relative production rate of fission product IRNs

IRN	Production rate of each IRN from each fissionable isotope divided by the average production rate of that IRN from all fissionable isotopes, %					
	U-233	**U-235**	**Pu-239**	**Pu-241**	**U-238**	**Th-232**
Thermal Neutron Spectrum						
Se-79	176%	55%	116%	54%	159%	41%
Tc-99	54%	58%	141%	147%	178%	22%
Sn-126	109%	26%	178%	86%	196%	4%
I-129	90%	35%	172%	103%	189%	11%
Cs-135	63%	52%	143%	142%	161%	39%
Average	98%	45%	150%	107%	176%	24%
Fast Neutron Spectrum						
Se-79	206%	80%	46%	68%	167%	33%
Tc-99	82%	97%	94%	126%	183%	17%
Sn-126	161%	65%	83%	92%	197%	3%
I-129	136%	71%	88%	105%	191%	9%
Cs-135	87%	90%	99%	123%	172%	28%
Average	135%	81%	82%	103%	182%	18%

3.3.2 Implications of Normalized IRN Production Rates

Observations on the comparison of differences in the IRN production rates among the six fissionable isotopes and three activation product precursor elements based on the results summarized in Table 6 and Table 7 are given in Table 8. When considering the observations, it is important to note that these are based on production rates for fissionable isotopes containing essentially only a single actinide isotope whereas real fuels can breed significant amounts of other fissionable isotopes during irradiation. The rationale for this analytical decision is found in Section 3.2. This assumption was necessary to attribute the IRN production rate to only one source. The next section will consider IRN production rates from fuels containing multiple precursor isotopes and elements.

Table 8. Observations concerning comparison of IRN production rates across fissionable isotopes

Observation Topic	Thermal	Fast	Comments
Fission product IRN production rates: fissile vs fertile FIs	Fertile/fissile production rates are <0.25%	Fertile/fissile production rates are <2.5%	U-238 and Th-232 fission cross sections are small so production rates are negligible compared to fissile isotopes and will be mostly ignored below.
Activation product IRN production rate variability	Variability of production rates across FIs SD ~5%: because essentially the same cross sections, fluxes and precursor concentrations were used High production rate variability among IRNs mainly due to cross section differences	IRNs ~ 23% smaller for U-233/Th due to lower flux Variability of production rates across FIs: SD 10-31% High variability among IRNs mainly due to cross section differences	Focus on activation product IRNs in this report will be minimized as they primarily depend on the precursor concentration and not the fissile isotope Concentrations of the stable precursors are poorly known; this constitutes a future R&D need
Comparison of fissile FIs: Normalized fission production rates for the average of 5 fission products and for I-129	Average: Pu-239 (150%) much higher. Pu-241 (107%) and U-233 (98%) about the same. U-235 much lower (45%) I-129: Pu-239 (172%) much higher. U-233 (90%) and Pu-241 (103%) about the same. U-235 (35%) much lower	Average: U-233 (135%) much higher. Pu-241 (103%) about the same. U-235 (81%) and Pu-239 (82%) lower. I-129: U-233 (135%) much higher, Pu-241 (105%) about the same, U-235 (71%) and Pu-239 (88%) lower	Relevant values are given in Table 7. I-129 was selected because it was found to be the dominant IRN in the preliminary study.

Table 8 (continued). Observations concerning comparison of IRN production rates across fissionable isotopes

Observation Topic	Thermal	Fast	Comments
Fission product IRN production rate variability for fissile FIs	Variability of production rates across fissile FIs: SD 43-54% due to yield differences between FIs The range of individual IRN production rates 30-300x mainly due to yield differences between IRNs	Variability of production rates across FIs: SD 14-64% The 60-300x range of production rate variability among IRNs mainly due to yield differences between IRNs	Mainly due to fission product yield differences among FIs
Si-32 production rate	The production rate of Si-32 is less than 10^{-6} of the next smallest IRN production rate	The production rate of Si-32 is less than 10^{-6} of the next smallest IRN production rate	Half-life of 153y Production rate is low because the nearest stable isotope is Si-30 and the half-life of Si-31 is just 2.6h plus the capture cross sections are very small (<0.1 barn)
IRN production rate dependence on burnup	The IRN production rates are essentially constant irradiation times of interest (<5y) because the mass increases linearly with burnup	Same	If irradiated long enough the production rate would decrease due to significant neutron captures by some IRNs.
IRN production rate dependence on precursor depletion	Depletion is about 4%	Depletion is 11-12%	The production rate of all IRNs will decline as their precursors are depleted by neutron capture or fission. No compensating adjustments were made.
Importance of bred fissionable isotopes	Preventing breeding by zeroing the capture cross sections of the FIs makes little difference for fissile isotopes but is important for fertile isotopes. When this measure was taken the fission product production rates were reduced by 6% to 20% for U-238 and by 10% to 32% for Th-232.	Preventing breeding by zeroing the capture cross sections of the FIs makes little difference for fissile isotopes but is important for fertile isotopes. When this measure was taken the fission product production rates were reduced by 1.5% to 4% for U-238 and by 1% to 25% for Th-232.	This effect is due to the small fission cross sections of U-238 and Th-232 coupled with a significant capture cross section that makes fissile isotopes having a large fission cross section

Key: IRN, important radionuclide; FI, fissionable isotope; SD, standard deviation

3.3.3 Production Rate of the IRNs from Representative Fuel Compositions

The previous section provided and discussed calculated IRN production rates from each of six single fissionable isotopes to provide a basis for determining the importance of various assumptions, parameters, and isotopes to IRN production. However, this approach does not represent real nuclear fuels which contain several fissionable and stable IRN precursor isotopes at the beginning of life (BOL) and many fissionable isotopes plus a few stable IRN precursors at the end-of-life (EOL). All of the precursors contribute to the production of IRNs to varying degrees depending on their initial concentration, fission product yields, fission and capture cross sections, etc.

This section takes the next logical step by describing calculations of IRN production rates for representative fuels containing multiple IRN precursors as a basis for comparing the integrated IRN production rates. The representative fuels are those identified in Table 10 for which ORIGEN2 models are available as described in the footnotes in Table 5.

The calculational approach is summarized as follows:

- The BOL fuel compositions and irradiation parameters were taken from the reference documents. The only change made was in the BOL composition of the PWR using U-233/Th fuel where the ORIGEN2 model assumed the U-233 was denatured with U-238 but instead in the model used for the follow-on study thorium was substituted for the fertile isotopes other than thorium in the BOL fuel. This was done to make the results less entangled by not having two fuel cycles (U-233/Th and LEU), and because the comparable fast reactor fuel did not contain any U-238. The four ORIGEN2 models were executed leading to four EOL fuel compositions. The BOL and EOL fuel compositions for the IRN precursors are shown in Table 9.
- The resulting IRN mass concentrations (g/MTIHM) were then transferred to a spreadsheet.
- BOL results will not be discussed because it is only the EOL IRNs that are relevant to repository risk assessment.
- The normalized IRN production rates (g/(MW_td-ppm) for each IRN fission product precursor in Table 6 were transferred to the spreadsheet.
- The mass concentration of each IRN (ppm) precursor was multiplied by the corresponding normalized IRN production rate to yield the production rate for each IRN (g/(MW_td) which is a suitable basis for comparing the four fuels.

The results from implementing the foregoing are summarized in Table 10.

Table 9. BOL and EOL fuel compositions for ORIGEN2 model reactor fuels (ppm)

Precursor	**Fuel Type**							
	BOL				**EOL**			
	PWR LEU	**PWR Th/U**	**SFR MOX**	**SFR Th/U**	**PWR LEU**	**PWR Th/U**	**SFR MOX**	**SFR Th/U**
N	25	25	25	25	25	25	25	25
Si	12	12	12	12	12	12	12	12
Cl	5	5	5	5	5	5	5	5
U-233	0	31425	0	172227	0	18717	0	124500
U-235	32000	2238	1674	4455	7936	2368	698	5652
Pu-239	0	0	128592	0	5033	2203	98520	0
Pu-241	0	0	5209	0	1223	589	12820	0
Th-232	0	966337	0	793115	0	941943	0	717300
U-238	968000	0	864525	30203	947059	0	777968.6	0.05
Total	1000042	1000042	1000042	1000042	961293	965862	890049	847495

Table 10. IRN production rate from two types of nuclear fuel in two neutron spectra

Fuel Type[a]	**IRN Production Rate, g/MW$_t$d and rank order of production rate (1 = smallest)**															
	Total		**C-14**		**Cl-36**		**Se-79**		**Tc-99**		**Sn-126**		**I-129**		**Cs-135**	
PWR-U	3.01E-02	1	4.11E-06	2	1.19E-05	4	1.38E-04	2	1.73E-02	3	8.33E-04	1	4.78E-03	1	7.04E-03	1
PTD3	3.62E-02	2	3.66E-06	1	1.05E-05	3	4.11E-04	3	1.86E-02	4	1.26E-03	2	7.41E-03	3	8.58E-03	2
AMORUUUC	5.78E-02	4	6.39E-06	3	3.86E-07	2	1.36E-04	1	1.70E-02	2	1.39E-03	3	5.87E-03	2	3.34E-02	4
AMO0TTTC	5.54E-02	3	4.94E-06	4	2.26E-07	1	4.95E-04	4	1.45E-02	1	2.15E-03	4	8.44E-03	4	2.98E-02	3
Statistics																
Average	4.49E-02		4.77E-06		5.74E-06		2.95E-04		1.68E-02		1.41E-03		6.62E-03		1.97E-02	
SD	1.20E-02		1.04E-06		5.46E-06		1.61E-04		1.47E-03		4.76E-04		1.40E-03		1.20E-02	
SD %	26.63%		21.77%		95.06%		54.48%		8.73%		33.78%		21.19%		60.82%	
Max/Min	1.9x		1.8x		52x		3.6x		1.3x		2.5x		1.8x		4.8x	
Rank[b] Thermal : Fast	Line Sum: 32 Line Sum: 38		3 7		7 3		5 5		7 3		3 7		4 6		3 7	
Rank[c] Pu/U:U-233/Th	Line Sum: 31 Line Sum: 39		5 5		6 4		3 7		5 5		4 6		3 7		5 5	

[a] PWR-U: PWR fueled with low-enriched uranium oxide; PTD3: PWR fueled with thorium enriched with U-233; AMORUUUC: SFR Pu/U core fuel; AMO0TTTC: SFR U-233/Th core fuel.

[b] Sum of rank for thermal (PWR-U+PTD3) and fast (AMORUUUC+AMO0TTTC) spectra. Smaller is better.

[c] Sum of rank for uranium fuel cycle (PWR-U + AMORUUUC) and thorium fuel cycle (PTD3 and AMO0TTTC). Smaller is better.

Observations concerning the comparative production rate of IRNs from the two fuel types in two neutron spectra based on the results in Table 10 are as follows:

- Mass IRN production rates do not account for differences in the rate of transport from the repository to the receptor or the application of factors to convert the mass received by the receptor to dose to the receptor. These differences are important because the preliminary study showed I-129 to be the dominant contributor to receptor dose whereas it is clearly not dominant when using production rate as a measure.
- The rank ordering (small to large) of the IRN production rates across the fuel/spectra combinations is not consistent.
- On average, the production rate increases in the order Se-79 < Sn-126 < I-129 < Tc-99 < Cs-135.
- The maximum/minimum of fission product IRN production rates ranges from 1.3x to 4.8x. This range is much less than the 30x-300x and 60x-300x ranges in Table 8 for individual fissile precursor isotopes and indicates that a mixture of fissile isotopes tends to ’average out’ the range. This would indicate that the choice of fuel type and neutron spectrum has a limited impact on the IRN production rates and, by extension, repository risk. The more important discriminator appears to be differences in subsurface transport rate and radionuclide-specific conversion of intake to dose (see the first item in this list).
- A qualitative measure of IRN production rate from thermal vs. fast neutron spectra and Pu/U vs. U-233/Th fuels can be developed by adding the rank for each of the five IRNs and summing them which is done in the bottom portion of Table 10. Remembering that a lower value is better, thermal vs. fast yields a score of 22:28 and uranium vs. thorium yields a score of 20:30. This suggests that a thermal reactor operating on a uranium-fuel cycle should have the lowest IRN production rate.

3.4 Conclusions and Recommendations

3.4.1 Conclusions

The major conclusions related to IRN production rates in thermal and fast neutron spectra from individual fissionable isotopes and representative nuclear fuel mixes are as follows:

1. The fraction of fissions that come from fertile isotopes is <0.25% for a thermal spectrum and <2.5% for a fast spectrum.
2. The activation product IRN production rates depend little on the spectrum or type of fuel. Instead, the production rates are heavily dependent on the concentration of stable precursor isotopes (typically trace levels) in the fuels and structural materials near the reactor core. In some cases (e.g., molten chloride salt reactors, reactors with nitride fuel), the precursor concentrations could be substantial. Trace-level activation product precursor concentrations are poorly known.

3. Fission product IRN production rates have a noticeable dependence on the type of fuel and the spectrum because of differing fission product yields and cross sections. However, the dependence is not systematic; production rates are higher for some fuels/spectra and lower for others.
4. Si-32 does not appear to be a likely candidate for addition to the list of IRNs because its production chain requires two neutron captures and the capture cross sections are small.
5. The depletion of activation product IRN precursors is essentially zero. The depletion of actinide isotope precursors can range up to 12% for high-burnup fast reactor fuels.

3.4.2 Recommendations

Based on the foregoing, the following are recommended:

1. Identify measurements of trace-level activation product precursors in fuels and structural materials in high-flux regions as an important R&D need.
2. Include activation product IRNs in the remainder of this study even if knowledge of precursor concentrations is incomplete.
3. Do not consider IRN production from fertile materials in the remainder of this study because the number of fissions in fertile materials is relatively small.
4. Do not consider Si-32 as an IRN in the remainder of this study because its production rate would be negligibly small even in reactors using silicon-based fuel or cladding materials such as SiC.
5. Do not consider the depletion of precursor isotopes in the remainder of this study because even the most extreme case (~100 GWd/MTIHM core fuel burnup) would lead to only a ~10% correction, which is negligible for the purposes of developing a SIRRAM.
6. The next steps in developing a SIRRAM should consider factors related to the rate at which IRNs are transported to the accessible environment and ingested by receptors leading to additional doses. These factors appear to be what leads to the significant differences in the importance of the IRNs.

4 UPDATE AND ANALYSIS OF THE REPOSITORY PERFORMANCE ASSESSMENTS UNDERPINNING A SIRRAM

The foregoing analyses are based on a set of IRNs identified in the preliminary study [Croff 2015]. This set of IRNs was culled from readily available PAs for the purpose of demonstrating that long-term repository risk depended on relatively few radionuclides. The preliminary set of IRNs provided some insights but was not intended to be exhaustive. With the demonstration having been made, it seemed necessary to address two additional issues which are the subject of this section:

1. Perform a wider search for repository PAs to increase confidence that no additional IRNs were identified in these analyses.
2. Attempt to ascertain the cause of the variability of IRN production rates from similar SNFs in similar host rocks.

4.1 Repository Performance Assessment Search Results

The results of an expanded literature search for repository PAs are summarized in Table 11. Note the following when examining this table:

- The search focused on documents that contained the results of PAs for mainly SNF although, in some cases, these documents also contain PAs for HLW.
- Only PAs that contained information that would allow numerical peak dose rates to be abstracted were considered. Several documents gave numerical values in a table for the peak IRN dose rates, but most contained the traditional time vs. dose rate color-coded curves for the contributing radionuclides.
- The PAs identified are all based on a ‘normal evolution’ scenario in which ground water eventually contacts and penetrates the waste package and cladding, dissolves the SNF matrix, and transports the radionuclides through the geosphere to the biosphere where it imparts a dose to humans.
- The tag given for the Preliminary Report analysis refers to PAs that were used in the preliminary study. If the tag is N/A, the associated PA was found as a result of the literature search performed for this analysis.
- “Country” refers to the location of the host rock analyzed and not necessarily the home of the analysts.
- Some of the references appear multiple times because they are reports that synthesize multiple PAs for multiple host rocks from multiple countries.
- The PA documents listed do not necessarily contain unique information. For example, there may be a full report, a presentation, and a summary paper all for the same repository host rock and design.

- The last column provides some key characteristics described as follows:
  - Dose rates: The location of the dose rate information in what are often large documents.
  - Time span: The length of time repository performance was analyzed.
  - Oxidizing: Whether the geochemical environment was oxidizing or not.
  - Waste type: The type(s) of waste considered in the PA.
  - Useful for SIRRAM: Whether the dose rates would be useful underpinning a SIRRAM.
  - New RN: A radionuclide that contributed to the SNF repository dose rate within one million years in a non-oxidizing environment, but which is not a previously identified IRN.

Table 11. Expanded literature review for repository performance assessments

PA #	Preliminary Report Analysis	Rock Type	Country	Reference	Comments
1	Granite-1	Granite	Spain	RED-IMPACT (2007)	• Dose rates: pdf 120 • Time span: 10MY • Oxidizing: No • Waste type: SNF • Useful for SIRRAM: Yes
2	Granite-2	Granite	Germany	Baudoin (2000) Gay (2000)	• Dose rates: Fig. 31 • Time span: 10MY • Oxidizing: No • Waste type: SNF • Useful for SIRRAM: Yes
3	Granite-3	Granite	USA	Mariner (2011)	• Dose rates: pdf 68 • Time span: 1MY • Oxidizing: No • Waste type: SNF • Useful for SIRRAM: Yes
4	Granite-4	Granite	USA	Mariner (2011)	• Dose rates: pdf 68 • Time span: 1MY • Oxidizing: No • Waste type: SNF • Useful for SIRRAM: Yes

Table 11 (continued). Expanded literature review for repository performance assessments

PA #	Preliminary Report Analysis	Rock Type	Country	Reference	Comments
5	Clay-1	Clay	Belgium	RED-IMPACT (2007)	• Dose rates: pdf 124 • Time span: 10MY • Oxidizing: No • Waste type: SNF • Useful for SIRRAM: Yes
6	Clay-2	Clay	France	RED-IMPACT (2007)	• Dose rates: pdf 126 • Time span: 10MY • Oxidizing: No • Waste type: SNF, HLW • Useful for SIRRAM: Yes
7	Clay-3	Clay	Germany	Rubel (2007)	• Dose rates: pdf 4 • Time span: 100MY • Oxidizing: No • Waste type: SNF • Useful for SIRRAM: Yes
8	Clay-4	Clay	Germany	Rubel (2007)	• Dose rates: pdf 4 • Time span: 100MY • Oxidizing: No • Waste type: SNF • Useful for SIRRAM: Yes
9	Clay-5	Clay	Belgium	Baudoin (2000) Gay (2000)	• Dose rates: Fig. 14 • Time span: 100MY • Oxidizing: No • Waste type: SNF, HLW • Useful for SIRRAM: Yes
10	Clay-6	Clay	France	Baudoin (2000) Gay (2000)	• Dose rates: Fig. 15 • Time span: 100MY • Oxidizing: No • Waste type: SNF, HLW • Useful for SIRRAM: Yes
11	Salt-1	Salt	USA	Swift (2012).	• Dose rates: pdf 26 • Time span: 1MY • Oxidizing: No • Waste type: SNF • Useful for SIRRAM: Yes

Table 11 (continued). Expanded literature review for repository performance assessments

PA #	Preliminary Report Analysis	Rock Type	Country	Reference	Comments
12	Salt-2	Salt	USA	Lee (2011)	• Dose rates: pdf 11 • Time span: 1MY • Oxidizing: No • Waste type: LWR SNF + DHLW • Useful for SIRRAM: Yes
13	Borehole-1	Borehole	USA	Freeze (2011)	• Dose rates: pdf 15 • Time span: 1MY • Oxidizing: No • Waste type: SNF • Useful for SIRRAM: Yes
14	Borehole-2	Borehole	USA	Swift (2012)	• Dose rates: pdf 20 • Time span: 1MY • Oxidizing: No • Waste type: SNF • Useful for SIRRAM: Yes
15	Borehole-3	Borehole	USA	Swift (2012)	• Dose rates: pdf 21 • Time span: 1MY • Oxidizing: No • Waste type: SNF • Useful for SIRRAM: Yes
16	Tuff-1	Tuff	USA	Swift (2019) Helton (2013) DOE (2008)	• Dose rates: pdf • Time span: 1MY • Oxidizing: Yes • Waste type: SNF • Useful for SIRRAM: Not unless Yucca Mountain licensing resumes. Outlier • New RNs: Pu-242 (30%), Np-237 (20%), Ra-226 (14%) of the peak dose sum

Table 11 (continued). Expanded literature review for repository performance assessments

PA #	Preliminary Report Analysis	Rock Type	Country	Reference	Comments
17	Tuff-2	Tuff	USA	Kozak (2009)	• Dose rates: pdf 228 • Time span: 1MY • Oxidizing: Yes • Waste type: • Useful for SIRRAM: Not unless Yucca Mountain licensing resumes. Outlier • New RNs: Np-237 decay chain (40%), U-234 decay chain (20%) of the peak dose sum
18	N/A	Tuff	USA	Van den Akker (2013)	• Dose rates: pdf 15, 16 • Time span: 100MY • Oxidizing: Yes • Waste type: HTGR hex graphite SNF • Useful for SIRRAM: Not unless Yucca Mountain licensing resumes. Outlier • New RNs: Np=237 (~80%), U-233 (~10%), Ac-227 (~5%) of the peak dose sum
19	N/A	Salt	Germany	RED-IMPACT (2007)	• Dose rates: pdf 128 • Time span: 10MY • Oxidizing: No • Waste type: SNF • Useful for SIRRAM: No; outlier. Based on altered scenario including anhydrite vein in salt, not normal evolution. See pdf 119 and Fig. 7.19 • New RNs: U-238 chain (~50%), Np-237 chain (~50%) of the peak dose sum
20	N/A	Granite	Canada	NWMO (2012)	• Dose rates: pdf 471 • Time span: 10MY • Oxidizing: No • Waste type: CANDU UOX SNF • Useful for SIRRAM: Yes • New RNs: Ni-59 and Ca-41 <0.1% of the peak dose sum

Table 11 (continued). Expanded literature review for repository performance assessments

PA #	Preliminary Report Analysis	Rock Type	Country	Reference	Comments
21	N/A	Granite	Canada	Chornoboy (2018)	• Dose rates: pdf 18 • Time span: 10MY • Oxidizing: No • Waste type: CANDU UOX and ThOX SNF • Useful for SIRRAM: Yes. Also has PA results for Be-10 • New RNs: Be-10 ~0.01% of the peak dose sum
22	N/A	Multiple	Japan	JNC (2000)	• Dose rates: pdf 205, 262, 309,318 • Time span: >1MY • Oxidizing: No • Waste type: HLW • Useful for SIRRAM: No. Focus is on HLW so there is no I-129. Outlier • New RNs: None
23	N/A	Clay	Switzerland	NAGRA (2002)	• Dose rates: pdf 261 • Time span: >10MY • Oxidizing: No • Waste type: SNF, HLW • Useful for SIRRAM: Yes • New RNs: None
24	N/A	Clay	Belgium	Marivoet (2009) NEA Report	• Dose rates: pdf 37 • Time span: 1MY • Oxidizing: No • Waste type: SNF, HLW • Useful for SIRRAM: Yes • New RNs: Pd-107, 0.001% of peak dose sum
25	N/A	Clay	Belgium	Van Iseghem (2009) NEA Report	• Dose rates: pdf 248 • Time span: 100MY • Oxidizing: No • Waste type: SNF PA result in background • Useful for SIRRAM: Yes • New RNs: Pd-107 (0.001%) and Th-229 (10^{-9}) of peak dose sum

Table 11 (continued). Expanded literature review for repository performance assessments

PA #	Preliminary Report Analysis	Rock Type	Country	Reference	Comments
26	N/A	Granite	Finland	Pastina (2008)	• Dose rates: pdf 101, 108 • Time span: 1MY • Oxidizing: No • Waste type: SNF • Useful for SIRRAM: Yes • New RNs: Nb-94 (0.001%), Pd-107 (0.001%), Ni-59 (0.01%) of peak dose sum
27	N/A	Granite	Sweden	SKB (2011)	• Dose rates: pdf 108, 120, 144 • Assumptions: Yes, detailed • Time span: 1MY • Oxidizing: No • Waste type: SNF • Useful for SIRRAM: Outlier due to assumptions • New RNs: Ra-226 (81%), Np-237 decay chain (10%) in a corrosion scenario and Ra-226 (~50%) and Ni-59 (~50%) in a shear scenario
28	N/A	Clay	France	ANDRA (2005)	• Dose rates: pdf 294, 315, 608 • Time span: 1MY • Oxidizing: No • Waste type: SNF, HLW • Useful for SIRRAM: Yes • New RNs: None. Only I-129, Cl-36, and Se-79 were shown over five orders of magnitude. Be-10 mentioned for alternative evolution scenario.
29	N/A	Clay	Switzerland	Zuidema (2021)	• Dose rates: pdf 8, 38, 47 • Time span: 10MY • Oxidizing: No • Waste type: SNF, HLW, ILW • Useful for SIRRAM: Yes • New RNs: None. Only I-129, Cl-36, C-14, and Se-79 contribute over four orders of magnitude.

Table 11 (continued). Expanded literature review for repository performance assessments

PA #	Preliminary Report Analysis	Rock Type	Country	Reference	Comments
30	N/A	Clay	Belgium	Chapman (2016)	• Dose rates: pdf 8 • Time span: 10MY • Oxidizing: No • Waste type: SNF • Useful for SIRRAM: Yes • New RNs: Ni-59 and Pd-107 each 10^{-6} of peak dose sum
31	N/A	Granite	Finland	Posiva (2012)	• Dose rates: pdf 231, 259 • Time span: 10MY • Oxidizing: No • Waste type: SNF • Useful for SIRRAM: Yes • New RNs: Ni-59 (0.1%) of peak dose sum
32	N/A	Clay	Netherlands	Baudoin (2000)	• Dose rates: pdf 77 • Time span: 100MY • Oxidizing: No • Waste type: • Useful for SIRRAM: No. No dose at less than 10MY.

4.2 Analysis of Repository Performance Assessments: IRN Identification

After conducting the preliminary study and the analysis of IRN production rates in Section 3, it was concluded that there were seven IRNs for non-oxidizing repository host rocks: C-14, Cl-36, Se-79, Tc-99, Sn-126, I-129, and Cs-135. However, after approximately doubling the number of PA results, as described immediately above, it is prudent to examine those results to see if any additional candidate IRNs were revealed. The results in terms of the frequency at which each IRN was identified are given in Table 12 and are discussed further below.

Table 12. Frequency of RNs in expanded PA literature review

Radionuclide	Preliminary Study		Follow-On Study		Outliers	
	Frequency	% of Total	Frequency	% of Total	Frequency	% of Total
C-14	6	11%	7	14%	1	3%
Cl-36	13	25%	9	18%	5	14%
Ca-41	1	2%	1	2%	0	0%
Ni-59	0	0%	3	6%	1	3%
Se-79	6	11%	7	14%	5	14%
Nb-94	0	0%	0	0%	1	3%
Tc-99	4	8%	3	6%	3	9%
Pd-107	1	2%	1	2%	0	0%
Sn-126	5	9%	4	8%	1	3%
I-129	15	28%	12	24%	5	14%
Cs-135	2	4%	4	8%	3	9%
Np-237	0	0%	0	0%	5	14%
Pu-242	0	0%	0	0%	5	14%
Total	53		51		35	

The frequency values in Table 12 are a count of how many times each IRN was included in the PA results. Additional assumptions are as follows:

- A 1-million-year time horizon was considered.
- The results are mostly for uranium-oxide-fuel LWRs although one result for a CANDU reactor is included.
- The results exclude radionuclides that contributed a small fraction (less than ~1%) of the total peak dose.
- The results are for SNF disposal in a non-oxidizing environment where the repository is undergoing normal evolution for its particular type of host rock.
- To enable a valid comparison, the results are separated into those identified in the preliminary study (see Section 1.1), those identified in this study (see Section 4.1), and the PAs that are outliers meaning their results cannot be meaningfully compared to the results from the preliminary or follow-on studies.

Observations based on the results in Table 12 are summarized as follows:

1. In general, the normalized frequency at which each radionuclide appears (percentages Table 12) in the preliminary and follow-on studies are essentially the same[4]. In particular,
    - I-129 is pervasively dominant.
    - There is a group of radionuclides that appear often: C-14, Cl-36 and Se-79.
    - There is a group of radionuclides that appear sometimes: Ni-59, Tc-99, Sn-126, and Cs-135.
    - The remainder of the radionuclides appear very infrequently.
2. When considering the results in Table 12, it is important to note that an IRN may appear less frequently because of differing assumptions concerning the amount initially in the SNF (e.g., different precursor concentrations), the rate at which the IRN is transported to the receptor, and even whether the set of radionuclides considered in a PA included all of those listed in Table 12.
3. Overall, the IRNs newly identified in the follow-on study confirm the set of IRNs adopted in both of the studies. The radionuclides in Table 12 that are not included appear sometimes or seldom and tend to contribute little to the total peak dose rate.
4. The rightmost portion of Table 12 contains the frequency at which the radionuclides appear in PAs deemed 'outliers', which means that the features of the PA differ sufficiently from the PAs considered in the preliminary and follow-on studies so that the results cannot be defensibly compared. Six of the 32 PAs fall into this category. The features that cause these PAs to be outliers are as follows:
    - Three of the PAs (Items #16, #17, and #18 in Table 11) are for an oxidizing repository host rock. The effects of an oxidizing host rock on radionuclide transport are substantial and are discussed detail in Section -1648147248. Briefly, in oxidizing conditions the actinide elements are much more mobile which makes the long-lived actinides and their decay products as important to risk as I-129, Cl-36, and Se-79 with none of them being dominant.
    - Two other outliers are the German salt PA (Items #19) and the Swedish granite PA (Item #27), where long-lived actinides and/or their decay products are substantial contributors to the dose rate.
        - In the case of the German salt repository, the evolution scenario selected was not a normal evolution and instead included an anhydrite vein that constitutes a path for rapid ground water transport of radionuclides that would ordinarily be substantially retarded by sorption.

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[4] Note that the frequency at which an IRN appears is not the same as its importance to risk because a particular IRN may only account for a small fraction of the total peak dose rate to a receptor.

    - In the Swedish repository, the granite is heavily fractured, and the receptor is located on the surface immediately above the repository which provides a path for rapid transport of radionuclides to the receptor. This is analogous to adopting a scenario that involves an anhydrite vein in the salt that provides a fast-path for water intrusion instead of the normal evolution repository scenario adopted in other PAs.
  - The last outlier is a Japanese PA (Item # 22) which assumed the high-level waste (HLW) was sent to the repository, not SNF. The HLW does not contain I-129 which brings the actinides and their decay products to the forefront.

4.3 Analysis of Variability Among Performance Assessment Results

The most useful features of PA results involve determining what constitutes an IRN and the dose rate of each IRN relative to the sum of the peak doses for all IRNs. For a given SNF composition, one of the most important causes of variability in both of these features is the redox condition in the geosphere between the repository and the biosphere. As is evident from the PAs in Table 11 that have oxidizing conditions (Items #16-18), the list of IRNs is dominated by actinides which is very different from for the rest of the PAs which do not have oxidizing conditions. This situation and its implications to a SIRRAM are discussed further in Section 5.3.

Another important cause of variability is rooted in the assumptions concerning the repository characteristics, especially those related to ground water ingress and migration to the biosphere. This is evident in Items #19 and #27 in Table 11, where the actinides are prominent. The reasons for this are discussed in Item #3 in Section 4.2 above.

Excluding the foregoing, the list of IRNs and the relative importance of the IRNs to repository risk is notably constant across the many PAs considered. Many radionuclides simply never exit into the geosphere because they are not very soluble, are strongly sorbed on the host rock, and/or the groundwater flow rate is sufficiently slow that they decay to stable isotopes first. The factors affecting the quantitative importance of the IRNs—which are long-lived and not strongly sorbed—are mainly characteristics of the ground water flow rate such as porosity, permeability, amount and nature of fracturing, and pressure gradient. Thus, there is an observed tendency for the identity and relative importance of the IRNs to vary relatively little; what changes is when the dose rate peaks occur. The next section considers these issues in the context of developing a SIRRAM.

5 DEVELOPING A SIRRAM

The foundation of a SIRRAM is essentially the process undertaken in Section 3. The first step is to use results from ORIGEN2, or equivalent, to calculate the normalized production rate (grams IRN per ppm precursor per MW_t) of each IRN (see Table 6). Next, the amounts of various nuclear materials (e.g., fuel, cladding) and the concentration of each IRN precursor in those materials are obtained. The normalized production rate and the IRN precursor concentration are multiplied, on an IRN-by-IRN basis, and the results are summed. The normalized IRN production rates are presented in Table 10 for four fuel type/neutron spectrum combinations. This result is a first-order measure of repository risk from a particular fuel cycle and reactor design, in that it is based on the calculated dose impact on a member of the public (as reflected in the PAs analyzed) to determine radionuclide importance. However, this first-order measure has a number of shortcomings. These shortcomings and the approach to alleviate them are discussed in the remainder of this section.

5.1 All IRNs Are Not Equally Important

The first-order measure accounts for the concentration of IRN precursors (stable isotopes or fissionable material) and the nuclear physics parameters involved in producing the IRN (e.g., fission yields, cross sections). However, simply adding the IRN production rates to yield a total mass of IRNs implicitly assumes that all IRNs contribute equally to receptor risk (dose rate) on a mass basis. To get to a measure of risk that is more representative of the actual risk, it is necessary to combine the IRN production rates with factors accounting for (a) the rate at which each IRN would be transported from a repository while undergoing radioactive decay and then ingested by a receptor, and (b) the dose conversion factor (DCF) which is the dose per unit intake for each IRN. All of these considerations are taken into account in the performance assessments but are not reflected in the simple calculation of IRN production rate in Section 3.

In fact, the quantitative information provides the basis for calculating an importance weighting factor that reflects the risk from each IRN. The weighting factor for an IRN from a single PA is simply the average of the peak values for each IRN. However, calculating the average is not entirely straightforward because the underlying PAs do not always present annual dose rates for the same set of radionuclides. It is unclear whether this is a result of a particular radionuclide being less than the smallest value on the ordinate or whether the performance analyst simply did not include that radionuclide in their analysis. In any case, it is necessary to decide how to calculate the importance weighting factors when IRNs have a variable number of data points and others being assumed to be zero. The adopted solution was to use two approaches. In the first, the average relative dose rate value for an IRN for which a dose rate was not given in the PA was assumed to be zero which was included in the averaging calculation to determine the weighting factor for that IRN. In the second approach, the average relative dose rate value for an IRN was based on averaging only the non-zero dose rates for that IRN. The resulting weighting factors are provided in Table 13.

Table 13. Relative importance weighting factors for IRNs

Important Radionuclide	Relative Risk Importance		Standard Deviation
	Averaged Over All PAs	Averaged Over Non-Zero PAs	
I-129	8.73E-01	8.73E-01	2.37E-01
Cl-36	1.39E-02	1.39E-02	1.93E-02
Cs-135	6.77E-03	5.07E-02	2.08E-02
C-14	6.13E-02	1.53E-01	2.05E-01
Se-79	2.80E-02	4.67E-02	5.33E-02
Sn-126	1.51E-02	5.67E-02	3.96E-02
Tc-99	7.99E-04	3.00E-03	2.17E-03

5.2 Reactor Thermal Efficiency

Ignoring the potential use of the thermal energy from a nuclear reactor to supply process heat, the purpose of a nuclear reactor is to produce electricity. The thermal efficiency (MW_e produced per MW_t) of a reactor can vary depending on the type of reactor or, more specifically, the reactor core outlet temperature, the heat sink temperature, and the power conversion technology (e.g., steam or Brayton cycle) used. A higher thermal efficiency will result in a lower IRN production rate per unit of electricity produced. The thermal efficiency of modern LWRs is about 35% whereas GEN IV advanced reactors project thermal efficiencies up to 50%. All other things being equal, such an increase in thermal efficiency would reduce IRN production rates by 30%. Accounting for differences in thermal efficiency is relatively straightforward: divide the results of the SIRRAM calculation described above by the thermal efficiency.

5.3 Oxidizing Repository and Groundwater Transport Conditions

All of the PAs analyzed in the course of preparing this report—except one—have involved host rock geochemistry that maintains reducing conditions along the IRN transport pathway for substantial distances. Under these conditions, the solubility of most radionuclides is low and sorption is high which results in the rate of transport through the geosphere being slow to the point that they do not reach the accessible environment within a million years or before they decay to stable species. There are some exceptions to this outcome, which results in the need to consider additional IRNs that can reach the accessible environment in quantities that require consideration in PAs. Most commonly, the first prerequisite for a radionuclide being important under reducing conditions is a long half-life (the shortest IRN half-life is 5700y). In addition, the solubility of the RN compound has to be relatively high compared to other RNs and sorption on the geology along the transport path has to be relatively poor. For example, the actinides typically have both low solubility and high sorption, so they are not IRNs under non-oxidizing conditions and a normal evolution scenario whereas the halides typically are the opposite and are therefore typically IRNs.

The one exception to the foregoing is the repository that was proposed to be located at Yucca Mountain, NV. The proposed repository is located above the water table and in oxidizing conditions. There are significant differences between the two examples in what constitutes an IRN. In particular, in the non-oxidizing environment, the actinides and their decay products do not make a noticeable contribution within a 10-million-year timeframe. In an oxidizing environment, however, some actinides and their decay products become noticeable contributors to calculated dose rate at about 50,000 years and become dominant contributors to the dose rate at the end of one million years with their contribution still increasing. The important actinide contributors in an oxidizing environment are from the Np-237 decay chain (Np-237 → U-233 → Th-229) and the U-238 decay chain (Pu-242 → U-238 → U-234 → Th-230 → Ra-226). Abstracting the peak dose rate (which occurs at one million years) yields the following list of IRNs and their contribution to the total dose rate: U-238 decay chain (47%), Np decay chain (32%), I-129 (14%), Cs-135 (3%), Sn-126 (2%), Tc-99 (2%), Se-79 (1%), and Cl-36 (0.04%).

Taking the new IRNs and their importance into account in a SIRRAM presents a substantial challenge. The obvious approach—simply adding U-238 and Np to the list of IRNs listed in Table 4—raises some issues:

- The proposed SIRRAM is based on IRN production rates from precursor stable elements or fissionable material. The U-238 in the fuel has no conceptual equivalent to a precursor as do the activation and fission products; it is included in the reactor by design.
- Most advanced reactors contain substantial amounts of U-238 in their initial and subsequent fuels. The minimum U-238 concentration would seem to result from the use of 20%-enriched HALEU or Pu-enriched MOX fuel while other reactor types may have higher concentrations. Adding U-238 to the SIRRAM calculation would result in a substantially larger value which (a) would be misleading if a reducing repository environment were being considered and (b) could lead to the conclusion that U-238 should not be in the reactor fuel which goes counter to the purpose and design requirements of a reactor.
- While neptunium is not currently planned to be part of the fresh fuel of any of the advanced reactors, its second-order precursor (U-235) is, and it is also produced by decay of Pu/Am-241 so its production is inevitable.

Despite these challenges, there is no conceptual reason why the SIRRAM concept could not be applied to an oxidizing repository with a couple of adjustments. First, U-238 and Np need to be added as IRNs. Second, a separate set of weighting factors reflecting the importance of each in an oxidizing repository would need to be developed and used. Third, the production rate of U-238 is simply the amount fed to the reactors as opposed to being produced by nuclear reactions. However, as discussed above, the usefulness of doing so is questionable since the U-238 is needed for the reactor to operate and the amount needed is determined mainly by reactor physics considerations.

5.4 Example SIRRAM Calculation

This section presents an example calculation of the SIRRAM described above. The previous discussion is expressed in the following equation:

$$\boldsymbol{SIRRAM} = \frac{\sum_i [\boldsymbol{PR_i} \cdot \boldsymbol{W_i}]}{\boldsymbol{E}} \qquad \text{Eq. 1}$$

where:

SIRRAM = Simple improved repository risk assessment measure for a reactor/fuel system

$\underline{i}$ = Index over the seven IRNs: C-14, Cl-36, Se-79, Tc-99, Sn-126, I-129, and Cs-135

PR_i = Production rate of IRN_i, grams per MW_td produced in the fuel

W_i = Relative risk importance of IRN_i

E = Thermal efficiency of the reactor/fuel system (MW_e/MW_t)

The PR_i parameter reflects the concentration of precursors in the fuel and could also be expressed as the product of the precursor concentration and the normalized production rates in Table 6. In essence, the SIRRAM is the sum of risk-weighted IRN production rates.

The example case will involve calculating a SIRRAM for each of the four reactor/fuel combinations in Table 10. The source of the necessary parameter values are as follows:

- The seven IRNs are identified in Table 4 excluding Si-32 (based on Recommendation 4) in Section 5.6.2.
- The production rate of each IRN for each fuel-spectrum combination is identified in Table 10.
- The relative risk importance values of each IRN are given in Table 13.
- The thermal efficiency is assumed to be 0.33 for PWRs and 0.40 [Vasile 2012] for SFRs.

5.5 Results of Example SIRRAM Calculation

The example calculations were performed in a spreadsheet and the relevant results were abstracted without further processing. The results are summarized in Table 14 and 15. Table 14 uses IRN risk importance obtained by averaging the peak dose rates for each IRN across all of the PAs assessed while Table 15 uses risk importance obtained by averaging over the non-zero peak dose rates (see Section 5.1).

The three sub-sections of Table 14 and 15 are described as follows:

1. The first sub-section contains the contribution of each IRN to the reactor type in the leftmost column and then the sum of these which is the SIRRAM in the rightmost column.
2. The second sub-section contains the fractional contribution of each IRN to the SIRRAM for each reactor type.

3. The third subsection contains (a) the sum of the two SIRRAMs for each IRN for the two SFRs divided by the sum of the two SIRRAMS for each IRN for the two PWRs (i.e., fast/thermal) and (b) a similar ratio for each IRN and the total SIRRAM for the two reactors using U/Pu fuel to the two reactors fueled with Th/U.

Table 14. Results of SIRRAM calculation for four reactor-fuel type combinations using all relative risk importance values of IRNs

Reactor Type[a]	**Important Radionuclide**							**Total SIRRAM or Ratio**
	C-14	**Cl-36**	**Se-79**	**Tc-99**	**Sn-126**	**I-129**	**Cs-135**	
	IRN contribution to SIRRAM (absolute)							
PWR-U	1.73E-07	5.00E-07	1.17E-05	4.20E-05	3.81E-05	1.26E-02	1.44E-04	1.29E-02
PTD3	1.54E-07	4.41E-07	3.48E-05	4.49E-05	5.78E-05	1.96E-02	1.76E-04	1.99E-02
AMORUUUC	2.22E-07	1.34E-08	9.51E-06	3.40E-05	5.25E-05	1.28E-02	5.66E-04	1.35E-02
AMO0TTTC	1.72E-07	7.84E-09	3.46E-05	2.90E-05	8.13E-05	1.84E-02	5.05E-04	1.91E-02
IRN contribution to SIRRAM (fractional)								
PWR-U	1.34E-05	3.88E-05	9.11E-04	3.26E-03	2.96E-03	9.82E-01	1.12E-02	
PTD3	7.74E-06	2.22E-05	1.75E-03	2.26E-03	2.90E-03	9.84E-01	8.84E-03	
AMORUUUC	1.65E-05	9.95E-07	7.06E-04	2.52E-03	3.89E-03	9.51E-01	4.20E-02	
AMO0TTTC	9.00E-06	4.11E-07	1.82E-03	1.52E-03	4.26E-03	9.66E-01	2.65E-02	
Ratio of SIRRAMs by reactor spectrum and fuel cycle								
SFR/PWR	1.20E+00	2.26E-02	9.48E-01	7.24E-01	1.39E+00	9.69E-01	3.34E+00	0.99
U/Th	1.22E+00	1.14E+00	3.03E-01	1.04E+00	6.51E-01	6.72E-01	1.05E+00	0.68

[a] PWR-U: PWR fueled with low-enriched uranium oxide; PTD3: PWR fueled with thorium enriched with U-233; AMORUUUC: SFR Pu/U core fuel; AMO0TTTC: SFR U-233/Th core fuel.

Table 15. Results of SIRRAM calculation for four reactor-fuel type combinations using relative non-zero average risk importance values of IRNs

Reactor Type[a]	Important Radionuclide							Total SIRRAM or Ratio
	C-14	Cl-36	Se-79	Tc-99	Sn-126	I-129	Cs-135	
	IRN contribution to SIRRAM (absolute)							
PWR-U	1.73E-07	5.00E-07	1.96E-05	1.58E-04	1.43E-04	1.26E-02	1.08E-03	1.40E-02
PTD3	1.54E-07	4.41E-07	5.81E-05	1.69E-04	2.17E-04	1.96E-02	1.32E-03	2.14E-02
AMORUUUC	2.22E-07	1.34E-08	1.59E-05	1.28E-04	1.97E-04	1.28E-02	4.24E-03	1.74E-02
AMO0TTTC	1.72E-07	7.84E-09	5.78E-05	1.09E-04	3.05E-04	1.84E-02	3.78E-03	2.27E-02
	IRN contribution to SIRRAM (fractional)							
PWR-U	1.34E-05	3.88E-05	9.11E-04	3.26E-03	2.96E-03	9.82E-01	1.12E-02	
PTD3	7.74E-06	2.22E-05	1.75E-03	2.26E-03	2.90E-03	9.84E-01	8.84E-03	
AMORUUUC	1.65E-05	7.71E-07	9.12E-04	7.33E-03	1.13E-02	7.37E-01	2.44E-01	
AMO0TTTC	9.00E-06	3.46E-07	2.55E-03	4.80E-03	1.35E-02	8.12E-01	1.67E-01	
	Ratio of SIRRAMs by reactor spectrum and fuel cycle							
Fast/Thermal	1.20E+00	2.26E-02	9.48E-01	7.24E-01	1.39E+00	9.69E-01	3.34E+00	0.98
U/Th	2.56E+00	1.16E+00	6.10E-01	1.69E+00	1.57E+00	1.30E+00	4.04E+00	0.71

5.6 Analysis of the SIRRAM Example Calculation Results

The following can be concluded by examining Table 14:

1. Iodine-129 is the most important IRN. It accounts for no less than 95% of the risk for all reactor types. Cesium-135 is the next most important, accounting for ~1-4% of the risk for all reactor types. The other three fission products are in the ~0.1-0.4% range. This is in contrast to the production rates of I-129 and Cs-135, where the latter slightly exceeds the former on average (see Table 6).
2. The activation products contribute essentially nothing to risk measure with the largest contribution being ~0.004%. However, this might not be the case for revolutionary fuels such as carbide, nitride, or chloride fuels containing unenriched anions. Again, it is important to remember that the production of fission product IRNs is controlled mainly by nuclear physics whereas the concentration of activation products is controlled by the concentration of stable precursor isotopes.
3. On average, fast reactors have a negligibly smaller SIRRAM than thermal reactors, and U/Pu fuels have a significantly smaller SIRRAM than Th/U-233 fuels.

The following can be concluded by comparing Tables 14 and 15:

1. Iodine-129 is still the most important IRN to risk. However, its fractional risk importance is diminished to 74-90% compared to using the relative risk importance averaged over all PAs. This occurs because I-129 imparts essentially the same dose to a receptor in all PAs whereas the other IRNs only do so in some PAs. When the other IRNs are averaged, the divisor is a smaller number which increases the relative risk importance of the other IRNs. This change brings Cs-135 into some prominence (~ 7-24%). The other IRNs are also increased but they were much less important than when averaging over all PAs, and thus the absolute increase is small.
2. The total SIRRAM is increased compared to averaging risk importance over all PAs, and this is also a direct result of the causation.
3. On average, fast reactors have a negligibly smaller SIRRAM than thermal reactors, and U/Pu fuels have a significantly smaller SIRRAM than Th/U-233 fuels.

6 ASSESSMENT OF THE UTILITY OF THE SIRRAM

At this point a SIRRAM has been developed and demonstrated by example. This section will address uncertainties and impediments to calculating the SIRRAM, and limitations in using the SIRRAM to discriminate among advanced reactor and nuclear fuel designs.

The purpose of developing the concept for a SIRRAM and performing an example calculation was to provide insights on how various advanced reactor and fuel choices might affect post-closure repository performance. In the course of developing the SIRRAM a number of insights have been obtained, some of which do not require a SIRRAM *per se*.

6.1 Insights from the Calculation of IRN Production Rates

Insights obtained during the research and analyses leading up to, but not including the SIRRAM, are:

1. A small number of radionuclides (IRN activation products: C-14 and Cl-36; IRN fission products: Se-79, Sn-126, Tc-99, I-129, and Cs-135) account for essentially all of the peak annual dose rate to a public receptor because they have relatively long half-lives and significant solubility even under non-oxidizing conditions and/or are retarded relatively little during transport.
2. A relatively small number of repository PAs are exceptions to the insight immediately above; they are outliers. In addition to oxidizing geochemistry, this results from the selection of repository evolution scenarios other than the normal evolution, specific features of the site being analyzed (e.g., fracturing, proximity to the dose receptor), or reprocessing the SNF which removes important radionuclides such as I-129. In such cases, the actinides and their decay products generally make substantial contributions to the peak dose rates.
3. For fission product IRNs:
   - The precursors of fission product IRNs are fissionable actinide isotopes. The mass production rate of IRNs per unit of energy produced varies significantly among the IRNs for various fissionable isotopes mainly because of differences in fission product yields but varies relatively little (standard deviations on the order of ±50%) across fissionable isotopes. Further, fertile isotopes which are fissionable produce negligible amounts of IRNs and can be ignored with little loss in accuracy.
   - The mass production rate of fission product IRNs from representative fuels where fissions occur in multiple fissionable isotopes vary less than the rates from single fissionable isotopes.

4. For activation product IRNs:
    - The precursors of activation products are isotopes of stable elements (N-14 for C-14 and Cl-35 for Cl-36). The concentration of these elements in fuels is not constrained by nuclear physics and can therefore vary significantly depending on design or methods of material manufacture. For most fuels, the activation product precursors of interest are trace contaminants present in small concentrations (tens of ppm maximum) which results in low IRN production rates.
    - For some advanced reactor fuels (nitrides, chlorides, and possibly carbides) the precursor concentration can be tens of thousands of ppm (or possibly over 100,000 ppm) which could make the resulting IRN production very significant to repository risk. For these reactors, this high precursor concentration is a liability; this fact is understood and has engendered continuing discussion of enriching nitrogen and chlorine to lower the concentration of the troublesome precursors.
    - The concentration of trace activation product IRN precursors in nuclear fuels and structural materials is not well known because the precursors do not impact other aspects of the nuclear fuel cycle and so are infrequently measured and/or published.
    - The neutron capture cross section of Cl-35 in a fast spectrum is also not well known Research to address this situation is ongoing [Terrapower 2022].
5. Thermal reactors and U/Pu fuel have smaller aggregate IRN production rates than fast reactors and Th/U fuel.

The insights from calculating IRN production rates need to be considered in the context of the uncertainties in the parameters and assumptions underlying the results. With respect to the fission products, the uncertainties are small because the production rates are normalized to energy production and the mass of fissionable material and consequently depend on well-known nuclear physics parameters such as fission product yields, cross sections, and half-lives. As discussed in Item #4, there are significant uncertainties in the concentration of activation product precursors and the capture cross sections in a fast neutron spectrum which could be important in reactor systems having high concentrations of Cl-35 or N-14.

There are also some conceptual limitations to obtaining insights from calculated production rates alone. The most important limitation is that simply comparing production rates implicitly assumes that the IRNs are transported through the geosphere and biosphere at the same rate, have the same specific activity (e.g., Ci/g), and the same activity-to-dose conversion factors. This is clearly not the case and is the reason for using a SIRRAM, which is discussed in the next section.

6.2 Additional Insights from the SIRRAM Calculation and Results

In Section 5, the conceptual limitations in using production rates by themselves were described and then addressed by weighting each IRN by a relative risk importance value obtained from evaluating existing PAs. In essence, this procedure is intended to address differences in the rate at which each IRN is transported to the receptor and the dose rate to the receptor.

Additional insights from the SIRRAM calculation and results are as follows:

1. The SIRRAM represents a distinct improvement over previous measures of repository risk from various radionuclides and nuclear fuels such as radioactivity, toxicity, an even IRN production rates because it accounts for the differentiating effects of radionuclide transport from the repository to the environment.
2. The magnitude of the SIRRAM component for each IRN that is summed to yield the total SIRRAM further emphasizes the dominant importance of I-129, with Cs-135 having decreased importance relative to I-129 compared to importance-based on production rate alone (see Section 5.6 Item #1), and the other IRNs being negligible.
3. The variability of the SIRRAMs for the four reactor/fuel types compared to production rates from single fissionable isotopes is further reduced so the standard deviation is 18% to 19%.
4. Insights on thermal vs. fast reactors and U/Pu vs. Th/U fuels are unchanged from Item #5, above.
5. While this follow-on study did analyze some thorium-based LWR and SFR fuels, and MOX SFR fuel, the analysis implicitly assumed that the set of IRNs and relative risk weighting factors for LWR oxide fuels was applicable to other fuels. This assumption could be characterized as a first approximation and it introduces additional uncertainties when used to compare the advantages and disadvantages of advanced nuclear reactor systems having significantly different mixes of fissile materials and SNF degradation rates in repository environments.
6. It might be possible to rely on just the PA results to identify IRNs and establish relative risk weighting factors for each IRN as a basis for comparing the relative repository risks from advanced nuclear fuels. However, doing so would implicitly assume that the IRN precursor concentrations were the same as those in the underlying PAs whereas use of the SIRRAM allows alternative precursor concentrations unique to advanced fuels to be reflected in the results.

These additional insights need to be considered in the context of the following limitations in the SIRRAM:

- As numerical peak dose rate values for individual radionuclides (e.g., from tables) are generally not available, they have to be extracted from graphs of dose rates as a function of time; as these are typically log-log graphs and low resolution, this introduces uncertainty.
- The set of radionuclides for which dose rates are provided is not the same in all of the PAs. It is unknown whether this is because the missing radionuclides are left out as a result of a screening process at the outset to reduce the calculational burden or whether their contribution to the total dose is less than the lowest value on the ordinate for some PAs. This limitation was addressed by examining two different approaches to determining the risk importance of the IRNs (see Table 14) with the result being that this limitation leads to inconsequential differences in the SIRRAM.
- The example SIRRAM was based on PAs for non-oxidizing repository environments. Except for the PAs for Yucca Mountain, NV, none of the other reference PAs addressed oxidizing repository environments. The list of IRNs for the Yucca Mountain site is very different than the list for a non-oxidizing environment; the calculated dose rate is dominated by the Pu-242 decay chain, the Np-237 decay chain, and then I-129.
- Minimizing the dose rates from these actinide decay chains would logically lead to a conclusion that the amount of U-235 and U-238 in a reactor should be minimized which would seem to be contrary to what is needed to operate current reactors and developing advanced reactors.

7 RECOMMENDATIONS

Based on the foregoing analysis, conclusions and discussion, the following recommendations are made:

- **Continue** to highlight the importance of the small set of radionuclides identified in this report to repository risk.
- Do not consider developing a SIRRAM for an oxidizing repository until/if efforts to license such a facility (e.g. Yucca Mountain) occur.
- **Encourage** more and better measurements of nitrogen and chlorine concentration in fuels and fuel assembly structural materials, along with the measurement of the neutron capture cross section of Cl-35 in a fast neutron spectrum, based on their roles in characterizing repository risk.
- **Consider** supporting a periodic effort to update:
  - Potential IRNs observed from newly published PAs.
  - The body of knowledge pertaining to the composition of planned advanced reactor fuel, cladding, and coolant materials.
- **Consider** focusing on the insights that can be obtained from calculation of IRN production rates in lieu of further development of the SIRRAM at this time, until uncertainties and limitations discussed in this report are addressed.
  - Fast reactors have a slightly smaller SIRRAM than thermal reactors.
  - U/Pu fuels have a significantly smaller SIRRAM than Th/U fuels.
- **Consider**, for follow-on work, a study that compares the IRNs derived from the SIRRAM to IRNs derived from the radiotoxicity metric and other commonly used measures of repository risk.

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