Nuclear Thermal Propulsion (NTP) Fission Product and Source Term Analysis

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DOI: ####

Nuclear thermal propulsion (NTP) is an in-space propulsion technology that uses a nuclear reactor to directly heat a propellant to provide high thrust, at higher efficiencies than achievable with conventional chemical propulsion systems. Due to the ability of NTP rockets to efficiently provide high thrust, this technology has been proposed for long duration space missions such as crewed missions to Mars and beyond. A primary hurdle for NTP technology development is the demonstration of an integrated reactor-engine system to verify its functionality and performance. This primarily requires the manufacture and testing of candidate reactor and engine technologies to demonstrate the proposed design enables desired performance and exhibits acceptable response under all known operation modes. A major component in this testing is fission product accumulation, since after operation radioactive isotopes can be produced and pose radiological dose concerns. This paper explains the background behind fission product analysis and the methodology used to evaluate current reactor designs and identify the amount of fission products present and their radiological impact. Results of multiple thrust level engines at different time periods are discussed and highlight the need for additional experimental testing to confirm fractional release levels of fission products out of the system. Benchmarking of models and future paths of analysis are also discussed.

I. INTRODUCTION

Prior to testing a reactor system, analysis must be performed to show that such testing will not result in radiological hazards or exposure to potential workers. Such analysis typically requires identifying the fission product inventory within the reactor due to its operation and performing a source term analysis. Source term, as defined by the Nuclear Regulatory Commission (NRC), is types and amounts of radioactive or hazardous material released to the environment following an accident. Therefore, quantifying exactly how much of a certain fission product is released by measuring the total production over the lifetime of the reactor is essential for risk mitigation and licensing decisions. This report discusses the reactor design used for the analysis and the processes used to find the fission products over different times of operation. These fission products are then compiled and converted to dose rates and compared to set occupational limits, followed by a discussion of these results on what fission products are of a higher concern than others for activities above dose limits. This source term analysis was performed at AMA in support of NASA's Space Nuclear Propulsion (SNP) project's NTP Ground Test Study.

I.A. Background

Fission products are the resulting atomic fragments that remain after a nucleus undergoes the fission process in a nuclear reactor. These products are usually unstable and will decay, leading to a release of radiation (typically beta or gamma decay), to reach a more stable state. The rate of decay is governed by the half-life of the isotope, which is the time required for the isotope concentration to decrease to one half its initial value. The formulas for three forms of exponential decay are shown in Equation 1 through 3 below.

$$N(t) = N_0\left(\frac{1}{2}\right)^{\frac{t}{t_{1/2}}} \tag{1}$$

$$N(t) = N_0\, e^{\frac{-t}{\tau}} \tag{2}$$

$$N(t) = N_0\, e^{-\lambda t} \tag{3}$$

N_0 is the initial quantity, N(t) is the remaining quantity after time t, $t_{1/2}$ is the half-life, τ is the mean lifetime, and λ is the decay constant, which all give the remaining quantity but use different methods to calculate it, depending on the given information. Half-lives are automatically compiled in reactor physics codes for the fission products accumulated, as their decay follows the half-life equation with their own specific decay constant. Fission product inventory is of interest to source term analysis because they can result in higher inherent reactivity during reactor handling or if released, they can be occupational hazards to any person in proximity. Short half-lives usually have high energy release and will result in higher doses until after the majority of their concentration has decayed, so using the fission product data to find the time after operation for safe handling is a valuable metric.

All fission products that emit radiation can be characterized based on their rate of decay, the decay products emitted during the decay process, and the harmfulness of the decay based on emitted particle and its energy. Activity characterizes the rate of decay of any radioactive isotope and has units of Becquerels (Bq), disintegrations/decays per second, which may be converted to Curies (Ci) which are 3.7e10 disintegrations per second. Activity measurements for different isotopes are important to compare the source term of the reactor against established limits and standards for facility safety and operations. While activity defines the rate at which ionizing radiation released, and different dose measurements can describe the impact of the decay of specific isotopes on the source term:

- ***Exposure*** is the amount of radiation traveling through the air, measured in roentgen (R)
- ***Absorbed Dose*** is the amount of radiation absorbed by an object or person. Absorbed dose has units of in *radiation absorbed dose* (rad) or the SI unit *Gray* (Gy), where 1 Gy = 100 rad.
- ***Dose Equivalent*** combines the amount of radiation absorbed with the medical effects of that kind of radiation. Dose equivalent has units of *roentgen equivalent man* (rem) and *sievert* (Sv)
- 1 R (exposure) = 1 rad (absorbed dose) = 1 rem or 1000 mrem (dose equivalent)

Dose definitions quantify the effects of the radiation from fission products in order to clearly assess the radiological risks and dangers associated with any handling or exposure near the reactor. Calculating these dose levels determine what levels of fission products can be deemed appropriate, hazardous, etc. More specifically, fission products need to be less than occupational dose limit standards set by government regulations. By finding the dose limits for proposed designs in an NTP testing environment the proper filtration and mitigation systems can be proposed and implemented for safety to workers and the public. Dose impact to workers is an important metric for this kind of analysis, and monitoring fission products from these proposed reactor designs is an important step for establishing the designs for testing and flight. By using reactor physics codes, the burnup of the reactor fuel can be simulated for whatever length of time is desired. Specific readers in the input decks can call out specific fission product masses to be reported in an output file. These fission product masses can then be converted into activity rates that can then be converted to dose rates and compared to occupational limits to see where the design sits. Fractional release can also be incorporated into the calculations. The fractional release refers to what percentage of fission products leak out of a system, so say 100% release would result in all accumulated fission products escaping the clad of the fuel and would release into the immediate vicinity. The analysis done in this study shows the highest possible activity levels for simple and conservative estimates, with the assumption that all products are released for maximum release potential. This study also considers NERVA legacy data that informs fractional release based on temperature-dependent diffusion data. These levels are still being discussed since test data on the diffusion of fission products available comes from NERVA data with different fuel designs than what is desired for new NTP mission parameters.

I.B. Methodology

Assessing the activity and dose of different fission products produced in a nuclear reactor needs several steps to properly calculate. An overview of the process is shown below in Figure 1, with a high-level description of the methodology followed by detailed discussion of the specific steps:

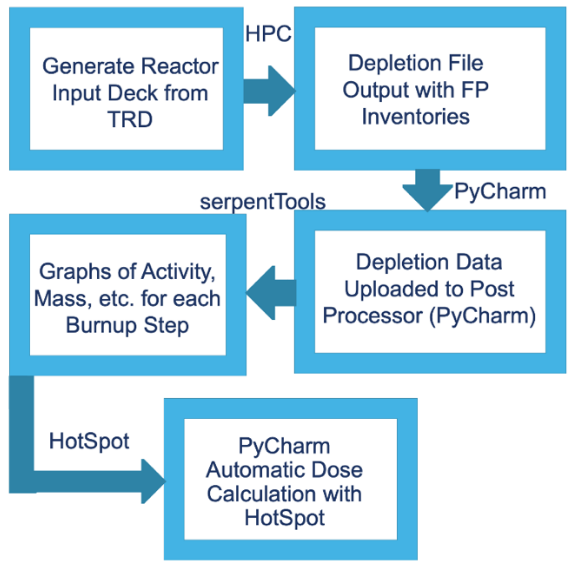

Figure 1. Analysis Pathway of Code Packages

Activity data is produced by the Serpent reactor physics model's depletion files. The code package serpentTools [1] was used to pull the relevant information by isotope name. All the helper scripts and packages developed were uploaded into a PyCharm interface that could interact directly with the HotSpot [7] source term code to provide specific doses and contour maps from a particular isotope. All activities had to be converted to Curies from Becquerels to work in HotSpot, then PyCharm takes those specific numbers and enters them into HotSpot. After HotSpot is run for that isotope, the Total Effective Dose Equivalent in rem was pulled from the output file and saved in PyCharm, which can then be plotted against other isotopes of note.

A reactor physics model was developed to accurately simulate the behavior of radioactive particles in one of the proposed designs for NTP applications. The Monte Carlo particle transport code used for this analysis was Serpent [6], which provided the burnup and inventory capabilities in a quick computational turnaround for test cases that were examined. The specific input files that were tested were based on the August 2020 Test Reference Design (TRD) specifications. The TRD inputs included Cermet and Cercer fuel forms and ranged from 12.5 klbf thrust levels to 25 klbf thrust levels to highlight the behavior of larger power systems fission product production compared to subscale systems. The reactor physics input files were set to burn the fuel materials and capture the fission product levels at all burnup steps defined in the scripts. These produced depletion files with the desired output data in activities of fission products for use in calculating dose.

The depletion and inventory were developed in a separate script that could be loaded into the main Serpent input file and edited to show different burnup steps and intervals depending on operation time, or what set of fission product inventories to display in the output. The Serpent inputs were run on the Idaho National Labs High Performance Computing cluster, with depletion files post processed with Python scripts developed in the PyCharm interface. The post processing scripts use depletion readers to pull all defined fission product activities from the output files and create Python arrays with the relevant data that can be copied and entered automatically with specific Python packages into HotSpot directly. The source term software HotSpot [7] developed by Lawrence Livermore National Labs was used to take input activity values and find dose estimates based on several input parameters that could be varied, such as release height, meteorological conditions, etc. which can be changed depending on given location weather impacts like ground shine and wind direction to carry the plume. For the scope of the analysis done with HotSpot, the initial conditions were kept to default values upon opening the code for meteorological conditions. The Material-At-Risk (MAR), which was replaced with the isotope activity of note, and the effective release height estimated to be 5 meters were the only modifications made to the default HotSpot parameters pending future site specific confirmations. The 5 meters was chosen based on the height of the potential release point of the plume on the proposed test rig design for ground testing.

The nature of the HotSpot interface requires a specific isotope to be named and its own activity entered into the MAR section in order to receive the Total Effective Dose Equivalent in rem in the output section. This quantity defined by the NRC is used to monitor exposure to ionizing radiation and used in NRC licensing documentation as a limit for many facilities and occupational documentation concerning radiological materials, and so is vital to calculate for ground testing activities. Hotspot can then create plume shapes and dose contour maps based on the release information input, but this process ran into the issue of being tedious to log all the values from a Serpent input even being post processed for activity. The PyCharm interface allows for a single script to take a depletion file from a Serpent run and output a series of specified isotope activities that are then fed into HotSpot automatically via Python calling functions and saved for easy data representation. For a scope of data needed in fission product analysis, leveraging these tools allow for efficiency and time savings in the broad scope of the analysis.

Fractional release was also a large focus of the fission product analysis. When the fuel in a nuclear reactor is heated and irradiated, fission products could migrate and diffuse through the cladding material and be released from the reactor fuel. Release of fission products can impact workers, filtration systems, and components in the overall system that could become irradiated. Understanding these phenomena and establishing initial estimates to be expected for the proposed designs is vital. Previous NERVA data referenced fractional release rates that were in the single digits of percent, only about 1% to 5% of the fission products generated were released. The NERVA designs tested were much different than current designs with much higher operating temperatures and fuel choices. Using the legacy data as a reference, estimated fractional release rates were compiled using WANL-TME-958 Interim Report on Fission Product Diffusion Code (FIPDIF) [5] and followed the equations below for derivation of fractional release:

$$1 - f = f_0 \, e^{-Dt} \tag{4}$$

$$D = D_0 \, e^{-E/(RT)} \tag{5}$$

In Equation 4 $(1 - f)$ is the fraction retained, f_0 is the fraction of diffusing species initially present which was set to 1, D is the apparent diffusion constant, and t is the time of operation. The rate constant is derived by Equation 5 where the activation energy (E) divided by the gas constant and the initial diffusion constant D_0 are given from Table 2 in the WANL report. Not all fission products were covered in the 7 diffusion groups presented in the table, but those that were had a specific compiled formula entered into the PyCharm interface. Nuclides analyzed in the WANL report included Sr-89, Ba-140, Ce-144, Y-91, La-140, I-131, Te-129, Cs-137, Mo-99, Zr-95, Ru-103. These temperature-dependent diffusion coefficient-based fractional release formulas are still dependent on Type II NERVA Fuel Elements and need to be updated with experimental data from new NTP designs. Once testing data of TRD fuel forms at proposed temperatures can be received and new diffusion data incorporated, the fractional release formulas can be updated for more accurate simulation. It is important to note that fractional release conditional statements were entered into the PyCharm interface and are used in the dose tabulations. Using Equations (4) and (5), for a specific group of isotopes, a temperature dependent release function is used to estimate the fractional release based on temperature.

II. RESULTS

Fission products can have a major impact on licensing and facilities decisions depending on what limits can be met where, and once the activities have been gathered from the Serpent input files, the results can be weighed against operational limits to inform these decisions. Hazard Category Analysis from the Department of Energy (DOE) is used to categorize nuclear facilities [2] [4]. The specifications of the different Hazard Categories are defined in Table 1 below.

TABLE I. Reproduction of Table 1 in Appendix A of Subpart B of 10 CFR Part 830 [4]

A DOE nuclear facility categorized as…	Has the potential for…
Hazard Category 1	Significant off-site consequences
Hazard Category 2	Significant on-site consequences beyond localized consequences
Hazard Category 3	Only local significant consequences
Below Hazard Category 3	Only consequences less than those that provide a basis for categorization as a hazard category 1, 2, or 3 nuclear facility

Assuming a full fractional release of all fission products present, the activities for both TRD Cercer and Cermet designs were run on the HPC, for the thrust ranges 12.5 klbf, 15 klbf, and 25 klbf. Burn times were varied over the course of the analysis for different examined results. Figure 2 showcases the list of all present fission products for the 12.5 klbf and 15 klbf Cercer and Cermet cases compared directly to Hazard Category Analysis (HCA) 2 limits imposed on the plot.

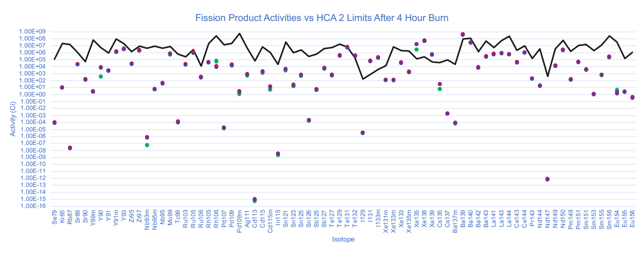

Figure 2. Fission Product Activities vs HCA 2 Limits After a 4 Hour Burn-time

All the fuel data after 4 hours of burn-time (the absolute maximum the reactor would run for over the lifetime of the design) had some isotopes running high and above HCA limits, but most falling under. The same comparison was done but for the reactor experiencing a 2-week cooldown after the 4-hour run time as shown in Figure 3.

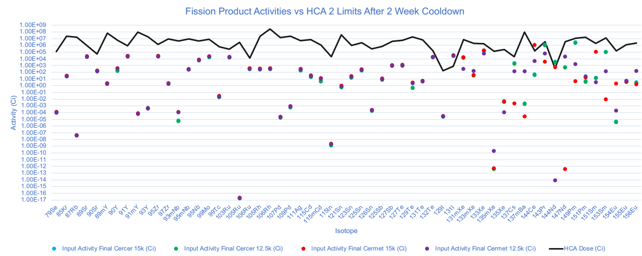

Figure 3. Fission Product Activities vs HCA 2 Limits After a 4 Hour Burn-time with 2 Week Cooldown.

Most of the isotopes decrease and stay under the limits, with the variation in fuel chemistry causing fluctuations in higher ZAI values and between fuel design choices. ZAI is a method of isotope identification using the atomic number (Z) followed by the atomic mass (A), so a ZAI value of 92235 would identify Uranium (92), and the 235 denotes the isotope of U-235. After these extended run times to show absolute maximum fission product accumulation, comparison between the Cermet and Cercer fuel forms suggests that Cercer has a more stable/consistent fission product behavior between the fuel cases. Only Xe-135 and I-131 are above the HCA 2 limit for the Cercer fuel, while I-131, Cs-135, La-140, Ce-141, and Pr-143 are above the HCA 2 limits for the Cermet Fuel.

The depletion method used in Serpent allows for time dependent behavior of isotopes to be modeled over time. This gives an insight into the decay behavior and can help determine if there are any isotopes acting conversely to what their half-lives would indicate. For example, I-131 is a high activity isotope that is of note for occupational hazard, and looking at its production over time in the 15 klbf Cercer test case in Figures 4 and 5 the operational behavior and shutdown behavior can be observed.

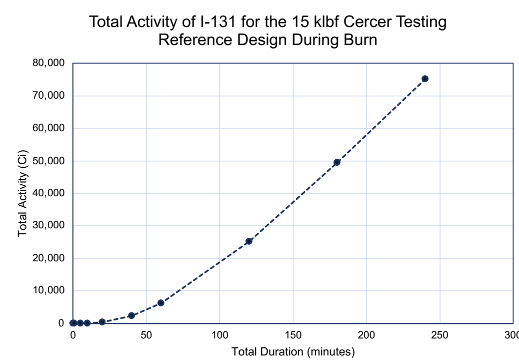

Figure 4. Total Activity of I-131 for 15 klbf Cercer Testing Reference Design During 4 Hour Burn

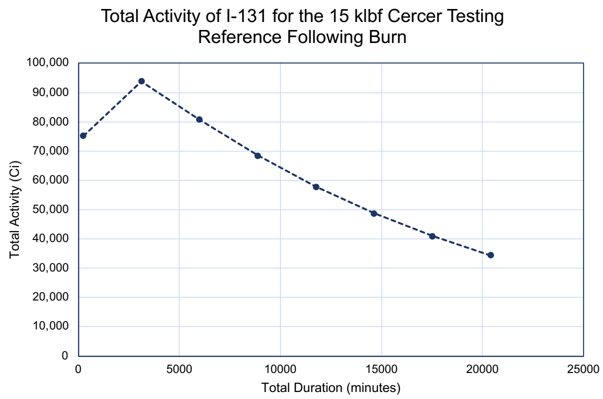

Figure 5. Total Activity of I-131 for 15 klbf Cercer Testing Reference Design During 2 Week Cool-Down

The isotope I-131 is highly volatile and radioactive, but since the half-life is about 8 days with a beta decay energy of about 0.1 MeV, it decreased quite quickly once cooldown begins. The steady decline from the shorter half-life will ensure that after a long period of time most of the I-131 will decay and not be a major hazard to workers, but proper shielding and handling analysis still needs to be examined. The nine highest activity isotopes as well as tritium for reference were plotted in the bar graph shown in Figure 6 with a black bar representing its specific HCA limit. The totals after 5 minutes of operation were shown for a more conservative level with a shorter run time more probable in ground testing, all thrust cases for Cercer shown as well as Cermet at 25 klbf for maximum values. I-135 is the only isotope over its limit, mitigation strategies will need to be discussed since most other isotopes fall below HCA limits.

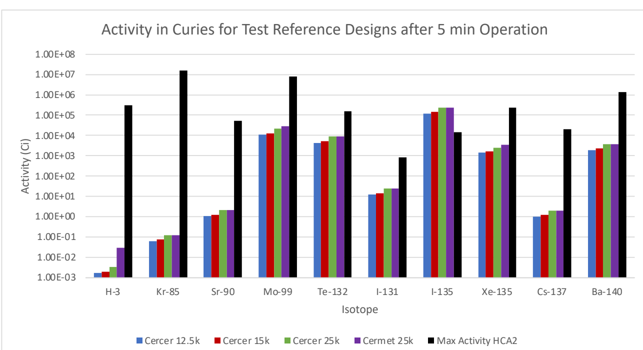

Figure 6. Total Activity in Curies for TRD After 5 Min Operation

With these total activity rates tabulated, the impact of fractional release needs to be quantified to see how much of a certain isotope can be released based on temperature. The range of diffusion coefficients used in the temperature dependent analysis are shown in Figure 7 below from the WANL FIPDIF report.

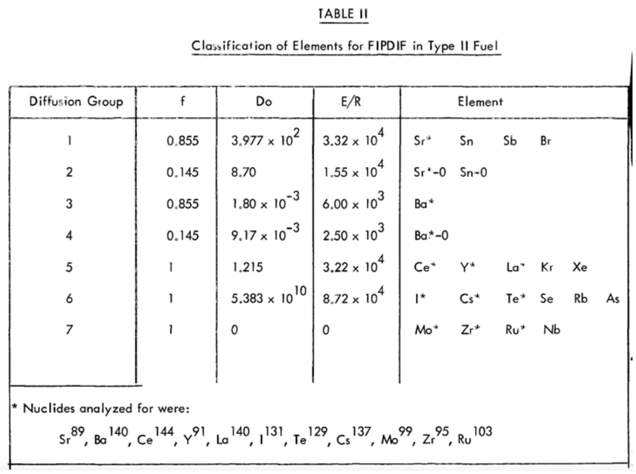
TABLE II

Classification of Elements for FIPDIF in Type II Fuel

Diffusion Group	f	Do	E/R	Element
1	0.855	3.977×10^{2}	3.32×10^{4}	Sr* Sn Sb Br
2	0.145	8.70	1.55×10^{4}	Sr*-0 Sn-0
3	0.855	1.80×10^{-3}	6.00×10^{3}	Ba*
4	0.145	9.17×10^{-3}	2.50×10^{3}	Ba*-0
5	1	1.215	3.22×10^{4}	Ce* Y* La* Kr Xe
6	1	5.383×10^{10}	8.72×10^{4}	I* Cs* Te* Se Rb As
7	1	0	0	Mo* Zr* Ru* Nb

* Nuclides analyzed for were:

Sr^{89}, Ba^{140}, Ce^{144}, Y^{91}, La^{140}, I^{131}, Te^{129}, Cs^{137}, Mo^{99}, Zr^{95}, Ru^{103}

Figure 7. Table 2 from Interim Report on FIPDIF [5]

In Figures 8, 9, and 10 the 12.5 klbf Cercer Serpent models were simulated for 5 min, 15 min, and 1 hour respectively with the fractional release dependent on the operating temperature implemented into the final dose values. As each activity at the final operation step was pulled from the Python scripts, it was multiplied by the associated group fractional release formula. For the results shown any element not included in the groups in Figure 7 was given a fractional release value of 0.6. The final dose values after automatic entering into HotSpot are shown below, the same set of elements displayed in Figure 6, and interposed with the Occupational Dose Limits for the General Public in a Controlled Area (0.1 rem per year, in Red) and General Employees (5 rem per year, in Green) [3].

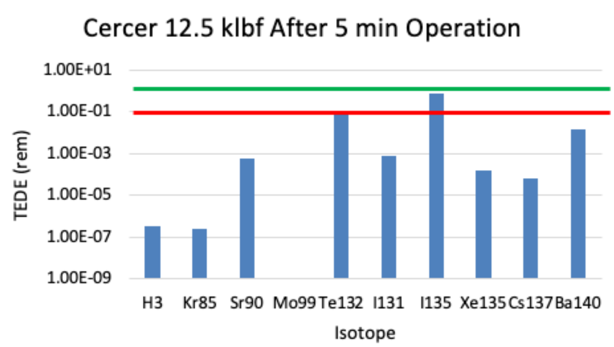

Figure 8. Total Effective Dose Equivalent of Highest Activity Isotope for the 12.5 klbf Cercer Case after 5 min

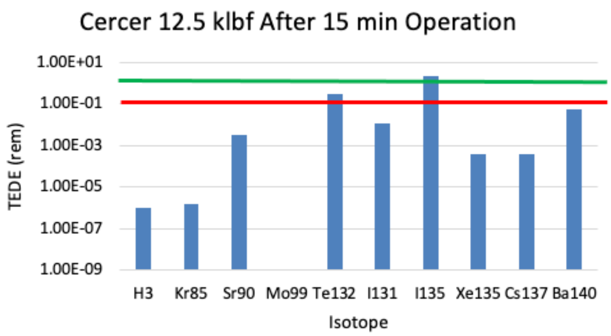

Figure 9. Total Effective Dose Equivalent of Highest Activity Isotope for the 12.5 klbf Cercer Case after 15 min

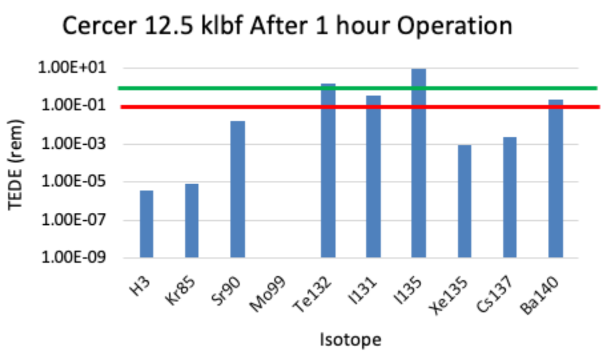

Figure 10. Total Effective Dose Equivalent of Highest Activity Isotope for the 12.5 klbf Cercer Case after 1 hour

In can be observed in these three final dose levels that the behavior of fission products over time meet expectations. The fractional release formulas for Mo-99 from the FIPDIF report result in a zero-percentage fractional release at the operating temperature of 2860 K. Isotopes like Te-132, I-135, and Ba-140 have much higher releases, almost 100% for some of them. With all these fission product inventories defined after 5 minutes, seeing the behavior over 15 minutes and then an hour, all products increase and reach closer to the Occupational Dose limits. For the 5-minute case, only Te-132 and I-135 are at or above the General Public in a controlled area level, while after an hour I-131 and Ba-140 exceed that limit as well. After an hour of operation Te-132 and I-135 exceed the Occupational Dose Limit for General Employees, indicating that the shorter run times for testing purposes these reactors undergo, the better dose limits can be achieved. This is pertinent to licensing decisions that will impact ground testing, as the design with the lowest amount of potential exposure and leakage of fission products will be the easiest to incorporate into any kind of testing proposal.

II.A. FUTURE WORK

The analysis presented in this paper is a stepping-stone to more complex burnup and source term analysis in support of an integrated NTP reactor-engine ground test demonstration. The dose levels of the fission products generated due to reactor operations can inform decisions about worker proximity, shielding requirements around the reactor, viability of open-air testing versus a filtration method, and to what extent of complexity the filtration system needs. Benchmarking to other source term analysis will also help refine the in-house modeling capabilities of the HotSpot Solver at AMA to be used for more expansive analysis in the future covering more areas of the source term automatically, such as implementing meteorological conditions, fractional release, and desired output data form all as user inputs. Continued support from subject matter experts at INL and the Nevada National Security Site will help inform proper assumptions and benchmark data in their own source term analysis. Shielding analysis for different material and cask designs will also be a major focus of work on the ground testing project. Activation analysis is also an area to examine, with more in-depth looks at particle interactions to provide for regulatory and licensing concerns.

III. CONCLUSIONS

Initial results indicate that the worst-case scenario of full release of all fission products will result in high dose rates that could reach the double digits in rem without proper shielding and mitigation. However, after incorporating temperature dependent fractional release formulas based on diffusion coefficients for these fission products, some isotopes will release fully or not at all, depending on their diffusion behavior. It is important to note however, the 12.5 klbf Cercer TRD case running for 5 minutes resulted in only one isotope, Iodine-135, being above the Occupational Dose Limit for the General Public in a Controlled Area (0.1 rem). With proper filtration systems in place fission products with high energy radioactivity can be managed without release into the environment. After weeks of cool-down, many fission products will also significantly decrease and allow for better handling. However, more research needs to be done on additional factors in the source term before definitive decisions can be made, such has site specific meteorological impacts on potential release.

ACKNOWLEDGEMENTS

This work was supported by NASA's Space Technology Mission Directorate (STMD) through the Space Nuclear Propulsion (SNP) project. This work was funded under Contract No. 80LARC17C0003. A special thanks to the subject matter experts at Analytical Mechanics Associates and Idaho National Labs for their assistance in this research. Another thanks to the Idaho National Labs High Performance Computing Cluster for the resources to complete these calculations. The serpentTools package on Github developed by the Georgia Institute of Technology's Computational Reactor Engineering (CORE) Lab was used to post process bulk depletion files quickly and accelerated the post run analysis capabilities by a significant amount, a special thanks to Dr. Kotlyar and the entire CORE Lab team.

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