

A FLEXIBLE MODEL FOR CALCULATING POTENTIAL FUTURE RADIONUCLIDE INVENTORIES

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The U.S. Nuclear Regulatory Commission (NRC) is developing a flexible performance assessment model, beta-Scoping of Options and Analyzing Risk (β -SOAR), to provide risk and performance insights on a range of permanent nuclear waste disposal options. One significant uncertainty in the β -SOAR pertains to the potential radionuclide inventory that will be disposed. The Inventory Process Model (IPM) was developed to calculate radionuclide inventories for a range of power plant operations; so that the inventories could be used in the β -SOAR.

The IPM calculates the radionuclide inventories based on external calculations using the ORIGEN-ARP component of the SCALE6 code system. A range of enrichment and burnup values were used with a fixed cooling time to generate 'databases' of radionuclide inventories. These databases could then be used, in conjunction with the number of operating reactors, uranium throughput and operational lifetime, to develop an initial radionuclide inventory. Additionally, the user may adjust the model to account for spent nuclear fuel, spent mixed-oxide fuel and high-level waste to provide a radionuclide inventory. These inventories could then be exported to β -SOAR to further enhance the model flexibility. An initial assessment of the model calculated total mass of the current U.S. inventory to within 5% of observed values.

I. INTRODUCTION

The NRC has developed a beta version of a flexible performance assessment, Scoping of Options and Analyzing Risk, Beta version (β -SOAR) to provide risk and performance insights for a range of potential geological disposal options for high-level waste and spent nuclear fuel. Five key components have been considered in the β -SOAR: the Waste Form, Waste Package, Near Field, Far Field, and Biosphere. Flexibility was a key consideration during development β -SOAR, and is described in more detail in an accompanying manuscript (Ref. 1). Currently, the β -SOAR has the capability to estimate the long term performance of three separate waste forms. The key considerations in the waste form component are the initial inventory and the degradation rate of each of the waste forms. Although the degradation

rates are waste form specific, the user has the flexibility to modify the inventories of the three different waste forms.

It is important to have an adequate understanding of what radionuclides are present because of the wide range of physic-chemical behaviors between them. For example, fission products such as ⁹⁹Tc and ¹²⁹I tend to be highly mobile in oxidizing environments. ⁹⁹Tc is redox-sensitive, with decreased mobility in reducing environments. These mobile radionuclides tend to be dominant early contributors to dose in performance assessments. Actinides tend to be less mobile than fission products, especially in reducing environments. When actinides do contribute to dose in performance assessment evaluations, they tend to lag behind the fission products.

There are three dominant classes of waste forms currently present in the U.S. The current U.S. high-level waste inventory is dominated by the spent fuel produced from pressurized water reactor (PWR) and boiling water reactor (BWR) commercial nuclear power plants. As of January 2010, the amount of commercial spent fuel stored at commercial nuclear power plants was an estimated at 63,000 metric tons [69,500 tons].² The amount of spent fuel is expected to increase by approximately 2,000 metric tons [2,200 tons] per year.² High-level waste from defense reprocessing is also present, and is represented as a vitrified, or glass, form in the β -SOAR model. Additionally, the DOE intends to utilize surplus weapons grade Plutonium as mixed-oxide fuel (MOX).² There are a number of operational factors that can impact the specific inventory for the waste forms, even within a given waste form class. For example, a spent nuclear fuel inventory can vary based on the burnup, initial uranium enrichment, reactor type and assembly.

To account for the variability in potential waste inventories, the Inventory Process Model (IPM) was developed to calculate the inventories from a wide range of potential fuel cycles. With respect to spent nuclear fuel, a number of PWR and BWR assemblies were considered. To enable the modeling of reprocessing scenarios, spent MOX fuel was also considered. The calculated inventories can then be exported to the β -SOAR to assess the effects of different waste form

inventories on performance in a range of geologic disposal systems.

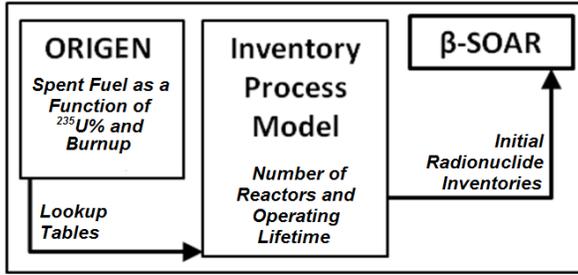


Fig 1: Flow of information in the Inventory Process Model

The IPM uses a suite of software: ORIGEN-ARP³ (ORIGEN) files, spreadsheets, automating scripts, and the GoldSim software. The IPM uses ORIGEN to generate databases that cover a broad range of potential radionuclide inventories. These databases are incorporated into a GoldSim model and allow the user to estimate the radionuclide inventory as a function of burnup, enrichment, assembly type, and reactor power level. To accomplish this, the values in the database, which were extracted from ORIGEN output files, can be interpolated. Each output file contains a radionuclide inventory produced from a particular set of parameters. The database of radionuclide inventories was generated by varying the conditions in each ORIGEN run and performing sufficient runs to cover the desired range of the parameters. This database can be used to represent the radionuclide inventory of spent fuel. Likewise, a similar database was generated for spent MOX fuel. A sufficient number of data points were developed in each database to interpolate without a significant loss of accuracy. The resultant inventories can be used to adjust the radionuclide inventories in the β -SOAR model. This paper discusses the development and the utilization of the IPM to develop the initial inventory of spent nuclear fuel for the β -SOAR.

II. MODEL DEVELOPMENT

The IPM uses ORIGEN calculated radionuclide inventories for spent fuel and spent MOX. Although ORIGEN can track the buildup of a large number of radionuclides in the spent fuel, the current version of the β -SOAR models a limited subset of radionuclides (Table 1). These radionuclides were selected for use in the β -SOAR because they exhibit a range of geochemical transport characteristics and are expected to show differences dependent on the fuel cycle assumed. Additional radionuclides were included in the databases produced by the IPM to maintain flexibility for future β -SOAR development, as needed.

TABLE 1. Radionuclides Tracked in the β -SOAR

¹⁴ C	⁷⁹ Se	⁹⁹ Tc	¹²⁹ I
¹³⁵ Cs	²³⁷ Np	²³² U	²³³ U
²³⁴ U	²³⁵ U	²³⁶ U	²³⁸ U
²³⁸ Pu	²³⁹ Pu	²⁴⁰ Pu	²⁴² Pu

Because precursors contribute significantly to the long term mass of ²³⁷Np, it was assumed that the ²³⁷Np precursors decay instantly. In other words to account for the decay of ²⁴¹Pu ($T_{1/2} = 14.4$ years) to ²⁴¹Am ($T_{1/2} = 432.2$ years) and then into ²³⁷Np ($T_{1/2} = 2.14 \times 10^6$ years) the initial amounts of ²⁴¹Pu and ²⁴¹Am from the ORIGEN output files are added to the initial amount of ²³⁷Np. Although ²⁴⁵Cm ($T_{1/2} = 8500$ years) is also a precursor to ²³⁷Np, it only occurs in small amounts in spent fuel and its contribution was assumed negligible. This addition of the main ²³⁷Np precursors provides a more realistic value for the long term ²³⁷Np inventory than using the initial values output by ORIGEN.

The IPM considered ranges for several of the ORIGEN input parameters: the reactor power level, assembly types, burnup, and initial ²³⁵U enrichments. The reactor power level was modeled as being either 20 or 50 Megawatts (MW)/basis. All ORIGEN calculations, for both UO₂ and MOX fuel, used 1 MTHM (both Uranium and Plutonium) as a reference basis. Four of the default assemblies ('w14x14', 'w15x15', 'w17x17', and 'ge10x10-8') from ORIGEN were used to capture the uncertainty in the radionuclide inventory caused by differences in assembly and reactor design. Assembly average burnups between 20 and 60 GWd/MTHM (gigaWatt-days per metric ton heavy metal) were evaluated at 2.5 GWd/MTHM increments. Initial enrichments between 2 and 6 wt% ²³⁵U were evaluated at 0.25 wt% increments.

The ORIGEN calculations included two moles of Oxygen for every one mole of Uranium or Plutonium in the fresh fuel composition. Modeling the Oxygen ensures that the ¹⁴C inventory resulting from the activation of Oxygen is calculated correctly. Nitrogen impurities in the fuel can also be activated to become ¹⁴C. To account for this, 100 g/MTHM [100 ppm] of Nitrogen was added to the starting composition of the fuels in the ORIGEN calculations. The Nitrogen impurity concentration in the UO₂ fuel matrix is limited to 75g/MTHM [ppm] by ASTM C776 Table 1.⁴ About 60% of the ¹⁴C is in the fuel;⁵ so this assumption should bound the expected ¹⁴C inventory which varies between 19 and 44 g/MTHM [ppm] in both the fuel and cladding.⁶ Other activation products (⁵⁹Ni, ⁶⁰Fe, etc) were ignored because the only activation product tracked in this version of the β -SOAR is ¹⁴C. Also note that the cladding and assembly hardware were not included.

For MOX fuel assemblies the Plutonium composition was assumed to come from the Plutonium composition of the spent fuel that was reprocessed. For example a fuel assembly with initial enrichment E may be burnt to a burnup of B; the Plutonium composition for that E-B pair may be used as the starting Plutonium composition for MOX fuel. However, ORIGEN will not accept Plutonium compositions with less than 50% or more than 70% ^{239}Pu (for example see Table D1.A.5 in Ref. 7 and Ref. 8); so spent fuel with Plutonium compositions outside this range are not modeled as being reprocessed. If the reactivity of the un-burnt MOX fuel is less than the reactivity of natural Uranium fuel enriched to 3 wt% ^{235}U then the fuel is assumed to not be reprocessed. The Uranium composition of the MOX fuel and the fraction of Plutonium in the MOX fuel can be controlled by the user when generating databases. A delay of 0.001 years was modeled between removing the spent fuel from the reactor and reprocessing. The delay between reprocessing the spent fuel, fabricating the MOX fuel, and loading the MOX fuel in a reactor was not modeled, so no Plutonium decay products were included in the fresh MOX compositions. To limit the number of ORIGEN calculations that have to be performed all spent MOX fuel is assumed to be burnt to 40 GWd/MTHM. Similarly, all the MOX fuel is represented by PWR MOX fuel.

The ORIGEN databases provide the inventory as a ratio of radionuclides on a MTHM basis as impacted by range of conditions described above. The databases are incorporated in the GoldSim model as lookup tables. When modeling reactors in GoldSim, an average yearly throughput of 20 MTHM of nuclear fuel per reactor was assumed. The number of reactors and operational period can also be modified by the user. The user can also determine the amount of spent MOX fuel to be included. The resultant SNF, HLW and spent MOX can be used to modify initial inventories in the β -SOAR model.

When modeling reprocessing in the IPM it was assumed that 100% of the Plutonium was removed from the spent fuel. The reprocessed Plutonium was then used as the starting point for the MOX fuel. The reprocessed Uranium was assumed to be separated and stored for future disposition. Since reprocessed Uranium might be disposed of separate from high-level waste, or re-enriched and used in UO_2 fuel, or used with reprocessed Plutonium in MOX fuel, or disposed of as HLW, the reprocessed Uranium and its decay products are tracked as a separate inventory. This will allow future versions of the β -SOAR to model the disposal of reprocessed Uranium if needed. The remaining material (fission products, higher actinides, etc) is considered high-level radioactive waste. Waste streams such as spent fuel cladding hulls are not modeled.

In the ORIGEN calculations the fuel was modeled as being continuously burnt at a constant power. ORIGEN calculations used relatively large time steps during irradiation, which results in minor errors in the spent fuel's radionuclide inventory due to the cross sections not being updated frequently. The differences between the radionuclide inventories produced from calculations with large and very small time steps were checked during scoping calculations and the error found to be only a few percent for the most sensitive radionuclides. Because this error is less than the uncertainty in other parameters, and that the β -SOAR is intended for scoping, the authors considered the error due to the time steps to be within acceptable levels.

Note that some of the ORIGEN database values may not be representative of the current spent nuclear fuel inventory. For example, inventory data was generated for 2.0 wt% enriched fuel burnt to 60 GWd/MTHM, although current nuclear power plants do not operate under these conditions. Likewise, low-burnup-high-enrichment cases would not be representative.

III. RESULTS AND DISCUSSION

III.A. Development of Representative Assemblies

III.A.1. Assembly Influence on Radionuclide Inventory

Seven PWR and seven BWR assemblies were assessed in the ORIGEN calculation. To assess the radionuclide inventory variability as a function of assembly, each assembly was modeled using an initial enrichment of 5 wt% ^{235}U and burnup of 60 GWd/MTHM. The assemblies modeled were the default assemblies available in ORIGEN. Variability of specific radionuclides between assemblies for a given reactor type (i.e. PWR, BWR) is generally within 5 to 10% (Fig 2 and 3). Note that only a subset of the radionuclides ultimately considered in the β -SOAR are shown, generally due to the extremely small contribution to total mass from the other radionuclides. In all cases the amount of ^{237}Np shown includes the amounts of ^{237}Np precursors.

Average PWR and BWR inventories were generated by using the results for the seven PWR and seven BWR assemblies shown in Fig 2 and Fig 3. The MOX inventories were generated by averaging the results for five PWR MOX assemblies from ORIGEN. The error bars highlight the maximum and minimum values calculated for each radionuclide. The MOX was modeled as being made from natural Uranium and 6 wt% reprocessed Plutonium. The reprocessed Plutonium was from the Plutonium composition of 5 wt% enriched fuel burnt to 40 GWd/MTHM. Although the range of radionuclide inventories for PWR and BWR assemblies

are within a factor of two, the difference between spent nuclear fuel and spent MOX is more pronounced (Fig. 4). On a per MTHM basis for the same burnup, spent MOX fuel has more Plutonium than spent UO₂ fuel. In general, for spent MOX there is a factor of two to five increase in plutonium and a factor of two to seven decrease in uranium (excluding ²³⁸U).

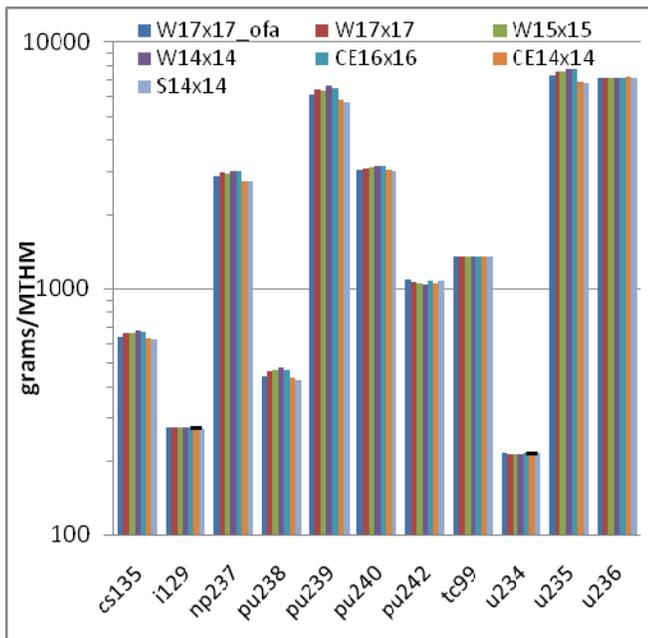


Fig 2: Scoping of select radionuclides for different PWR assemblies.

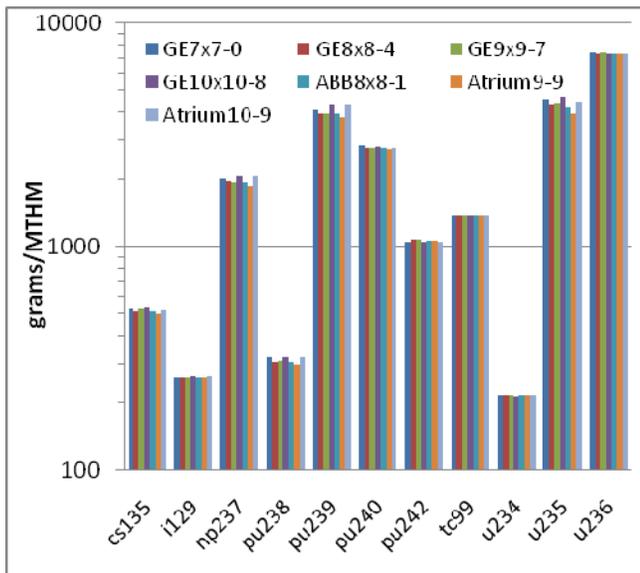


Fig 3: Scoping of select radionuclides for different BWR assemblies.

The similarity in resulting radionuclide inventories as a function of assembly type is also evident as a function

of burnup and initial enrichment (Fig 5 through Fig 8). In general there is less than a factor of two difference between all the PWR and BWR assemblies assessed. Additionally, there are regular trends observed for the assemblies as a function of enrichment and burnup. The similarity of the trends for BWR and PWR radionuclides is consistent with the trends observed in Fig 2 through Fig 4. The good agreement between the different assemblies shows that it is reasonable to use a representative or an averaged fuel in the β -SOAR, or other performance assessment code, to model the radionuclide inventory of similar spent nuclear fuels.

The spent MOX radionuclide inventory is dependent on the starting composition (i.e., the fuel being reprocessed) and fraction of initial Plutonium. The higher the initial loading of Plutonium the more Plutonium will be in the spent MOX radionuclide inventory. There is also a greater amount of ²³⁷Np in spent MOX fuel, which is important due to its long half-life (2.14×10^6 years); which means it will be available for release for a very long time. Neptunium-237 is more mobile in oxidizing environments than in reducing environments. MOX made from spent fuel that has been reprocessed very soon after being removed from the reactor will have more ²⁴¹Pu than MOX made from spent fuel that was allowed time to cool before reprocessing. Similarly, MOX fuel that was not immediately loaded into a reactor will have more ²⁴¹Am in its initial composition. When irradiated in a reactor this fuel will produce slightly more minor actinides due to neutron absorption by ²⁴¹Am. However, these changes are less significant than the burnup in the reactor and can also be impacted if there are long decay times before the fuel is available for release. Note that while Fig 5 through Fig 8 show PWR and BWR fuel assemblies, many of the trends will be similar, especially for fission products.

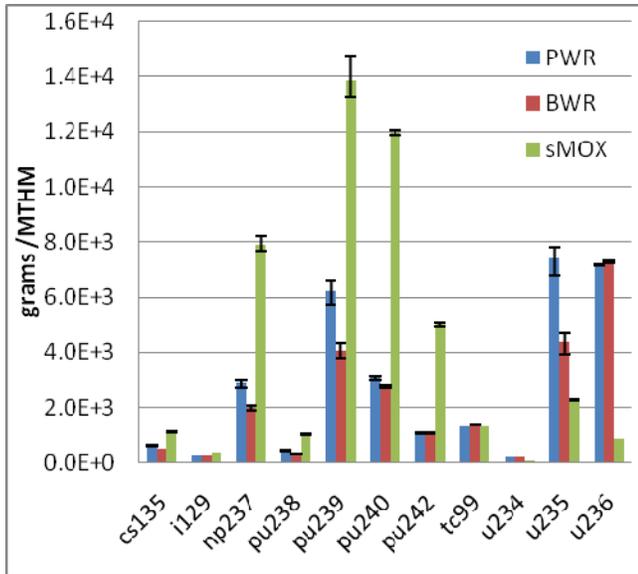


Fig 4: Radionuclide inventories for 60 GWd/MTHM burnt fuels. Ranges indicate the maximum and minimum values for seven PWR assemblies, seven BWR assemblies and five sMOX assemblies.

III.A.2. Burnup and Enrichment Influence on Radionuclide Inventory

A general trend was observed where higher burnup resulted in more fission products, and was observed in both spent UO_2 fuel and spent MOX. For a given enrichment, the trend is approximately linear as shown by the nearly constant slopes in Fig 5 and Fig 6. From Fig 4 it can be seen that the fission product inventory is relatively insensitive to the type and class of assembly. However, there are other factors affecting the radionuclide inventory such as the radionuclide undergoing fission. Each fissile radionuclide (^{235}U , ^{239}Pu , etc) will produce different fission product yields. For example the ^{129}I fission yield from ^{239}Pu is almost twice as large as its yield from ^{235}U ; so more will be produced at low enrichment because more ^{239}Pu is contributing to fission (at low to medium burnups). This effect can be seen in Fig 5.

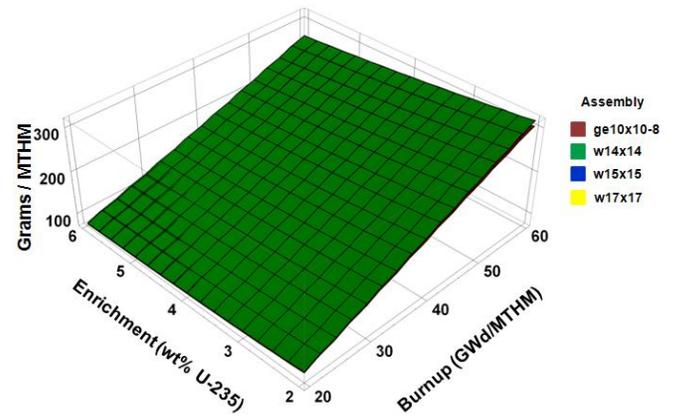


Fig 5: The ^{129}I inventory based upon a 1 MTHM feed basis, after 5 years of decay.

Another factor is the absorption cross section of a radionuclide and its precursors. For example, ^{135}Cs is primarily produced from beta decay of ^{135}Xe , which is a very strong neutron absorber (~ 3 million barns). At lower enrichments more ^{135}Xe absorbs neutrons and becomes ^{136}Xe , instead of decaying to ^{135}Cs . Because the overall fission cross section is higher at higher enrichments, more of the neutrons would be absorbed by ^{235}U instead of being absorbed by ^{135}Xe . This results in more ^{135}Cs at high enrichments as can be seen in Fig 6. This same phenomenon applies to other fission product absorbers.

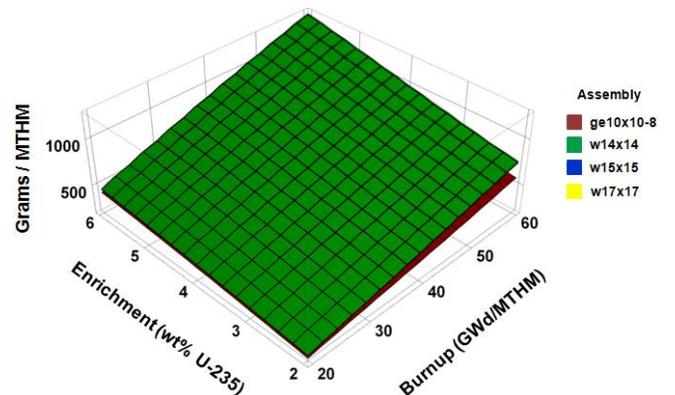


Fig 6: The ^{135}Cs inventory based upon a 1 MTHM feed basis, after 5 years of decay.

Another trend was observed with increased burnup; an increase of long-lived actinides. The relationship between burnup and the actinide concentrations is not linear. This is because many actinides have large neutron cross-sections and will either fission (Ex: ^{239}Pu) or transmute into a higher actinide (Ex: ^{240}Pu to ^{241}Pu). For example this effect will cause the rate at which ^{239}Pu builds up to decrease as burnup increases. As a fraction of the total Plutonium, the amount of ^{239}Pu will actually decrease with burnup. Additionally, many of the actinides are decay products of higher actinides. Some of

the actinide decay products that are tracked in the β -SOAR include ^{237}Np , ^{233}U , and ^{235}U . Fig 7 and Fig 8 show the difference between the ^{237}Np inventory that is present at 5 years and the ^{237}Np combined with its precursors which have much shorter half-lives. The ^{237}Np inventory shown in Fig 8 would be utilized by β -SOAR to account for the effect of ^{237}Np 's precursors on the long-term inventory by adding their 5 year inventories to the ^{237}Np inventory.

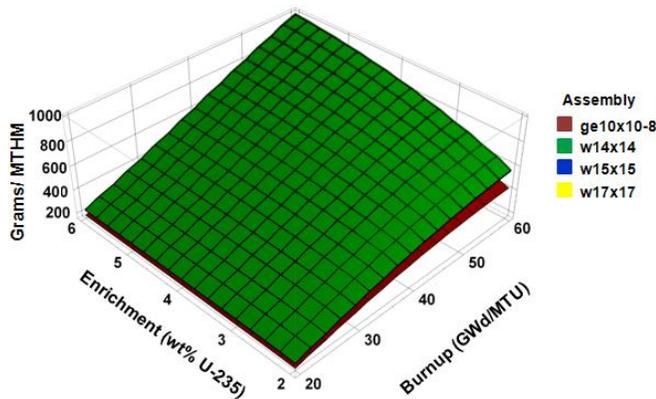


Fig 7: The ^{237}Np inventory based upon a 1 MTHM feed basis, after 5 years of decay.

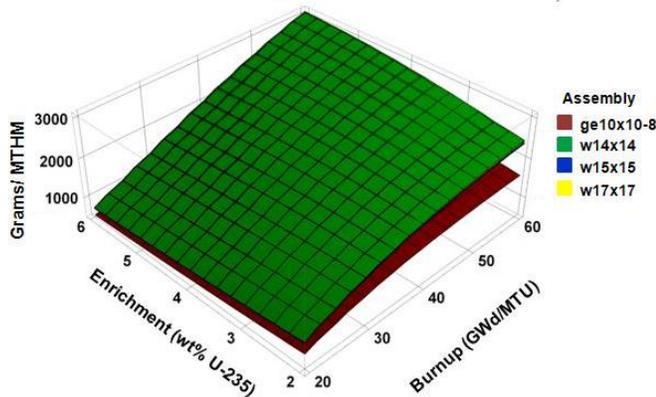


Fig 8: The ^{237}Np inventory combined with its precursors based upon a 1 MTHM feed basis, after 5 years of decay.

Although ^{14}C occurs in small amounts in spent fuel (i.e., a couple grams per MTHM or g/MTHM at most), and emits a weak beta particle (0.156 MeV) it is quite mobile and when present in the biosphere is readily absorbed into living organisms. Additionally, some or most of the carbon may be released in a gaseous form that would be even more mobile. In fuel ^{14}C is mostly produced via activation of Nitrogen impurities in the fuel, cladding, and hardware. For a fixed amount of Nitrogen in the fuel the amount of ^{14}C increases with increasing burnup, and decreases with increasing enrichment. This change as a function of enrichment is likely due to the Nitrogen absorption cross section becoming relatively more important as the overall fission cross section

decreases with decreasing enrichment. Although ^{14}C in the cladding and hardware would likely exhibit similar trends, the release rate would be different.

Because almost all the ^{14}C results from Nitrogen activation, the ^{14}C inventory can be scaled to approximate lower levels of Nitrogen impurities. For example the predicted ^{14}C inventory may be cut in half to approximate the results of 50 g/MTHM [ppm] of Nitrogen, instead of the 100 g/MTHM [ppm] modeled.

III.B. Initial Spent Nuclear Fuel Inventory for β -SOAR

The IPM was used to calculate the initial spent nuclear fuel inventory for the β -SOAR. The length and number of operating nuclear reactors was based on major operating reactors (~400 MWE) up through the end of the year 2010 with a 20 MTHM/yr throughput. Initial ^{235}U enrichment and burnup was assumed to be 5% and 40 GWd/MTHM, respectively. The resultant radionuclide inventory (Fig 9) is consistent with information available in the literature. The total radionuclide mass calculated by the IPM is approximately 67,800 metric tons [74,800 tons] at the end of 2010, which is within 5% of the estimated 65,000 metric tons [71,650 tons] at the end of 2010 (Ref. 2).

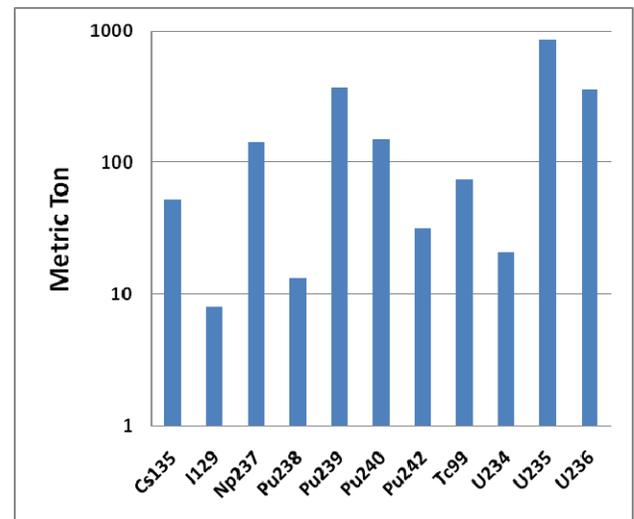


Fig 9: Initial spent nuclear fuel inventory in the β -SOAR

The calculations for the IPM indicate that it appears to be reasonable to use representative assemblies for spent nuclear fuel and spent MOX in calculating initial radionuclide inventories for β -SOAR. Values for individual radionuclides were shown to have a difference of only a factor of two when comparing different assemblies, although most were generally less than that. Additionally, this consistency was observed for the tested range of ^{235}U enrichment and burnup. In a demonstration

of the IPM, it was shown that the total mass of the spent nuclear fuel inventory at the end of 2010 appeared to be within 5% of the estimated value.

ACKNOWLEDGMENTS

This paper was a joint effort between U.S. NRC and CNWRA staff under contract NRC 02-07-006-000. The NRC and CNWRA staff views expressed herein are preliminary and do not constitute a final judgment or determination of the matters addressed or of the acceptability of any licensing action that may be under consideration at the NRC.

In addition the authors would like to thank Tae Ahn, Robert Einziger, and Zhian Li for their advice and comments.

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