Analysis of DOE Spent Nuclear Fuels for Repository Disposal

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Abstract – *U.S. Department of Energy (DOE) spent nuclear fuel (SNF) consists of hundreds of different fuel types in various conditions. In order to analyze and model the DOE SNF for its suitability for repository disposal, several generalizations and simplifications were necessary. This paper describes the methodology used to arrive at a suitable DOE SNF surrogate and summarizes the proposed analysis of this DOE SNF surrogate for its appropriateness as a representative SNF.*

I. INTRODUCTION

During the last four decades, the U.S. Department of Energy (DOE) and its preceding agencies generated approximately 250 varieties of spent nuclear fuel (SNF) from weapons production, nuclear propulsion, and research missions. Because many of these fuels were generated through research, a wide variety of fuels was developed. Not much is known about some of these fuels and some are in very poor condition. Information about all of the known DOE SNF is stored in a centralized database.[1] There are currently 622 SNF records in the database. It would be extremely cumbersome, if not impossible, to model each DOE SNF separately for repository performance assessment. It was necessary to develop an appropriate DOE surrogate fuel that would be representative of all existing DOE SNF. Methods were employed to group these varieties of DOE SNF and incorporate all the SNFs into one surrogate fuel. This methodology is described in this paper.

II. DEVELOPMENT OF A DOE SNF SURROGATE

An appropriate surrogate DOE SNF was developed in steps over several years. The following subsections summarize how this surrogate was derived.

II.A. DOE SNF Groups for Performance Assessment

The main goal of grouping the DOE SNF was to supply data in a cost-effective manner to support DOE SNF management and disposal without increased risk to the public, environment, or worker safety. A method was employed to group these varieties of DOE SNF into 11 categories for the repository performance assessment.[2] Each of the DOE fuels was grouped according to their fuel parameters. These parameters were determined based on properties that affect three major areas of licensing analysis: criticality, design basis events, and performance assessment. The parameters used to group the DOE SNFs are shown in Table I.

TABLE I. SNF Grouping Parameters

DOE SNF Parameter	Reason for using parameter
Fuel Matrix	Affects chemical reactivity, air oxidation rate, free radionuclide inventory fraction, gap fraction, and dissolution rate.
Fuel Cladding	Affects release over time and containment.
Fuel Condition	Affects particulates in fuel, release fraction, and surface area of fuel.
Fissile Species and Enrichment	Affects criticality and radionuclide inventory.
Reactor and Fuel Design	Affects radionuclide inventory.
Burnup	Affects radionuclide inventory and thermal effects.

The grouping methodology categorized the characteristics of a select number of fuel types by either a bounding parameter or by representing a particular characteristic of the whole category. Using the parameters listed in Table I, the entire range of DOE SNF was condensed into 34 intermediate groups. These 34 groups are listed below:

1. U Metal, Zr Clad, Disrupted, (LEU), N-Reactor
2. U Metal, Al Clad, (LEU), Single Pass Reactor
3. U-Zr, (HEU), CP-5 & HWCTR
4. U-Mo, Zr Clad, (HEU), Shippingport PWR
5. U Oxide, Zr Clad, Intact, HEU, Shippingport PWR
6. U Oxide, Zr Clad, Intact, (MEU), Saxton

7. U Oxide, Zr Clad, Intact, (LEU), Commercial
8. U Oxide, SST Clad, Intact, (HEU), ML-1
9. U Oxide, SST Clad, Intact, (MEU), PBF
10. U Oxide, SST Clad, Intact, (LEU), FFTF-TFA
11. U Oxide Failed or Declad, (HEU), SM-1A
12. U Oxide Failed or Declad, (MEU), ORNL SST & Zr
13. U Oxide Failed or Declad, (LEU), TMI-2
14. U Oxide, Al Clad, (HEU), HFIR
15. U Oxide, Al Clad, (MEU), FRR, MTR
16. U-Al or U-Alx, Al Clad, (HEU), ATR
17. U-Al or U-Alx, Al Clad, (MEU), FRR, MTR
18. U-Si, Al Clad, (HEU, MEU), FRR, MTR
19. U/Th Carbide, Graphite, Hi-Integrity, (HEU), FSV
20. U/Th Carbide, Graphite, Low-Integrity, (HEU), PB
21. U or U/Pu Carbide, Non-Graphite, (MEU, FGE), SRE
22. MOX, Zr Clad, (HEU, FGE), GE Test
23. MOX, SST, (HEU, FGE), FFTF-DFA
24. MOX, Misc Clad, (MEU&LEU FGE), FFTF-TFA-ACO
25. U/Th Oxide, Zr Clad, (HEU FGE), LWBR
26. U/Th Oxide, SST Clad, (HEU FGE), Dresden
27. U-Zr-Hx, SST/Incoloy Clad, (HEU), TRIGA Flip
28. U-Zr-Hx, SST/Incoloy Clad, (MEU), TRIGA Std
29. U-Zr-Hx, Al Clad, (MEU), TRIGA Alum
30. U-Zr-Hx, De-Clad, (HEU), TRIGA SNAP
31. Na-Bonded,SST/Misc,(HEU MEU&LEU),FERMI I Blkt
32. Classified, (HEU), Navy
33. Canyon Stab., (HEU & LEU), SRS Target
34. Misc. SNF,(HEU, MEU, & LEU), Misc, Unknown

After considering all of the parameters and properties important for post-closure performance, these 34 groups were then further grouped into 11 categories for performance assessment purposes. Naval fuel was placed into its own group (Category 1) for several reasons including: the design of naval SNF is significantly different than other DOE SNF designs (a very robust design), and naval SNF is classified.

Each DOE SNF fits into one of the following 11 performance assessment categories[3]:

Category 1	Naval Fuel
Category 2	Plutonium/Uranium Alloy Fuel
Category 3	Plutonium/Uranium - Carbide Fuel
Category 4	MOX (a mixture of U and Pu oxides)
Category 5	Thorium/Uranium - Carbide Fuel
Category 6	Thorium/Uranium - Oxide Fuel
Category 7	Uranium - Metal Fuel
Category 8	Uranium - Oxide Fuel
Category 9	Aluminum Based Fuel
Category 10	Miscellaneous Fuel
Category 11	Uranium - Zirconium Hydride Fuel

II.B. DOE SNF Radionuclide Inventories

Radionuclide inventories for SNF are necessary for performing any performance assessment calculations. An estimate of the radionuclide inventory for each DOE SNF was developed using a template methodology.[4] This template methodology enables radionuclide inventories to be estimated for virtually any SNF.

The methodology calculates radionuclide inventories for a typical spent fuel in each of the groups using reactor physics computer code, thus creating a template for the spent fuel group. The radionuclide inventory of each individual SNF can then be estimated by scaling the matching template by the mass and burnup of the individual SNF. The methodology uses a consistent process of using existing information, supplemented by conservative assumptions and estimates for missing information, to enable radionuclide inventories to be estimated for every DOE SNF.

These radionuclide inventories (for each DOE SNF) were then used to calculate an overall surrogate radionuclide inventory (in grams per waste package) by using a weighted average based on how each of the fuels would fit in canisters and be loaded into waste packages.[5] The resulting DOE SNF surrogate inventory is shown in Figure 1 along with a comparison to the inventories of each of DOE SNF categories.

Figure 1 illustrates that even though a specific category (such as Category 10) has a high inventory compared to the other categories, it has a small effect on the surrogate inventory because it makes up only about 0.1% of the total number of waste packages. The number of waste packages of each DOE SNF category is shown in Table II, along with the percentage of the total DOE SNF waste packages.

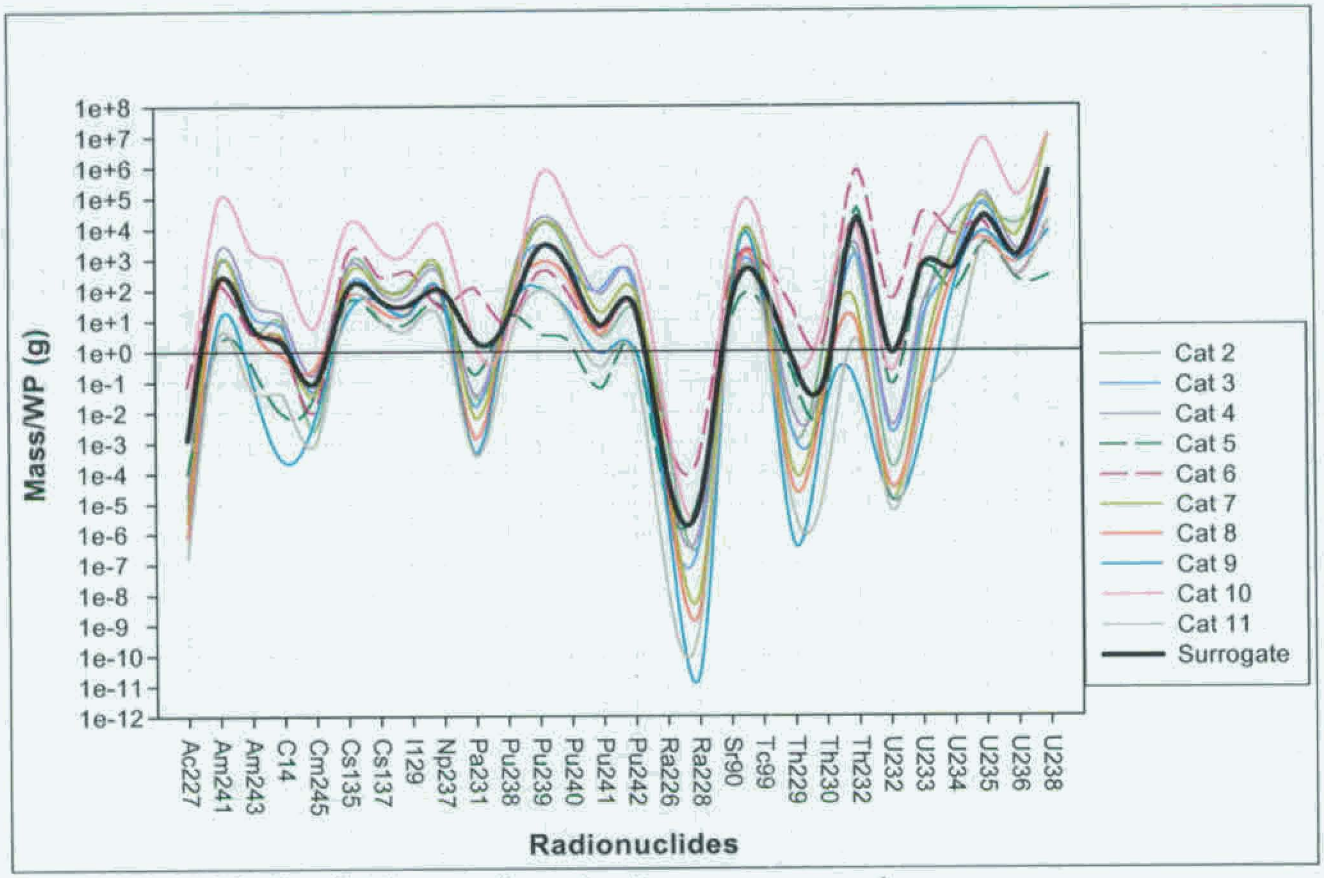

Figure 1. DOE SNF Radionuclide Inventory Comparison in grams per waste package.

II.C. DOE SNF Degradation Rates

The degradation rate selected to be used for the surrogate DOE SNF (for performance assessment) was instantaneous degradation, based on detailed degradation analysis for each of the DOE SNF categories previously mentioned (in Section II.A.).

Using the SNF dissolution equations developed in Reference 3, a fractional degradation rate was calculated for each of the DOE SNF Categories. These calculated rates are shown in Figure 2. A pH of 6.5 and a temperature of 95°C were used in the equations to calculate the degradation rate. The result of the calculation is the fraction of the fuel that is degraded in one year. If the calculated degradation rate is greater than or equal to 1, then the spent fuel is complete degraded in one year (instantaneous degradation). This occurs for categories 2, 3, 5, 7, and 10. These categories make up about 23.9% of the DOE SNF by waste package (volume).

Categories 8 and 9 contain a large amount of DOE SNF by volume (67.2%). Based on their calculated degradation rates, Category 8 DOE SNF degrades in about 6 years, and Category 9 DOE SNF degrades in about 18 years. In the context of 10,000 years this is essentially instantaneous. Category 4 DOE SNF has a very slow degradation rate but it makes up only 4.3% of the DOE SNF by volume. Category 6 DOE SNF is calculated to degrade in about 400 years and is only 1.9% of the volume. Category 11 DOE SNF is calculated to degrade in about 57 years and is only 2.8% of the volume.

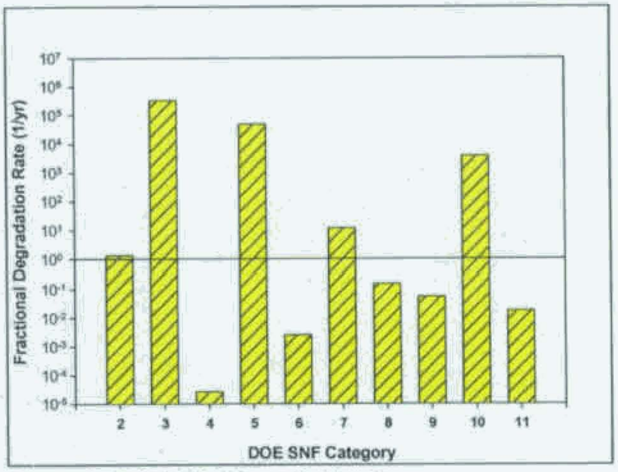

Figure 2. DOE SNF Degradation Rates
(pH = 6.5 T = 95°C).

Given that most of the DOE SNF degrades rapidly, it is justified to use instantaneous degradation for the DOE SNF surrogate. Also, this is recommended because it would be conservative and bound all the DOE SNF including those fuels in poor condition.[6]

III. ANALYSIS OF THE DOE SNF SURROGATE

The next logical step is to analyze the DOE SNF surrogate to determine if the derived DOE SNF surrogate is a reasonable representation of the entire range of DOE SNF. This section outlines how it is proposed that this is to be accomplished.

III.A. Recommended Methodology

To test to see if the DOE SNF surrogate reasonably represents each of the major categories of SNF, each of the individual DOE SNF categories needs to be run in a repository performance assessment model and compared to the surrogate. This should be done by performing a model run with one waste package of DOE surrogate failing (with no other waste forms present) and examining the expected dose result. Next, the same run needs to be performed by substituting the radionuclide inventory from Category 2 DOE SNF and using the specific degradation rate[3] for Category 2 (instead of instantaneous). The results then need to be compared with the surrogate. The same process should be followed for the rest of the DOE SNF categories (3-11). Naval fuel should be analyzed separately and compared to zircaloy clad commercial. Also as a final comparison, a weighted average (weighted based on the number of waste packages of each category) of the results from each category should be compared to the surrogate results. Table II shows the weighting factors that should be used for each DOE SNF category.

TABLE II. Number of DOE SNF Waste Packages

Category	Waste Packages	Percent of Total DOE SNF Waste Packages
Category 2	21	0.6 %
Category 3	11	0.3 %
Category 4	146	4.3 %
Category 5	569	16.7 %
Category 6	64	1.9 %
Category 7	211	6.2 %
Category 8	875	25.6 %
Category 9	1421	41.6 %
Category 10	3	0.1 %
Category 11	94	2.8 %

ACKNOWLEDGMENTS

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