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THERMAL MANAGEMENT OF WASTES FROM ADVANCED FUEL CYCLES

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I. INTRODUCTION

Alternative fuel cycles currently being considered would significantly change the overall heat output of HLW and/or directly disposed used fuel, compared to the direct disposal of once-through used fuel that has been U.S. policy since 1984 (Ref. 1). Heat output for U-Pu based fuel cycles is dominated by just a few radionuclides: short-lived fission products ^{137}Cs and ^{90}Sr, and transuranics (TRUs) with longer half-lives, principally ^{241}Am and several isotopes of Pu. In this paper we consider three fuel cycle cases that are idealized bounding states for scoping evaluations of repository temperature and waste radiotoxicity. These cases are: 1) use decay storage or other means prior to disposal of used fuel, to eliminate short-lived fission products; 2) burn TRUs present in once-through used fuel; and 3) continuously recycle TRUs with the objective to transmute and fission all the actinides including the ^{238}U present initially (e.g., in the current inventory of used fuel). In the results presented below, all three cases are evaluated with 100 years of additional decay storage (or active pre-closure repository ventilation) after 2067.

II. WASTE HEAT OUTPUT

For this study, reference used nuclear fuel (UNF) is represented using data from the Yucca Mountain performance assessment[2] projected to the year 2067 (nominally 50 years out-of-reactor). Three bounding cases were developed for analysis (Table I). Case 1 is the limiting state for eliminating heat generated by short-lived ^{137}Cs and ^{90}Sr, thereby decreasing the peak near-field repository temperature within a few tens of years after emplacement. Case 2 extracts additional fission energy from TRUs in used fuel, and eliminates the Am and Pu isotopes that are principal contributors to heat output between a few hundred years and 10,000 yr. This is the time interval when peak far-field temperatures occur for typical repository media and design concepts. For scoping analysis we assume perfect chemical separations and complete fission of TRUs, neither of which could readily be achieved in practice. Case 3 is an idealized example of full recycle in which actinides with mass number $\geq$ 232 are converted to fission products.

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Case 1 is simply the used nuclear fuel (UNF) without ^{137}Cs and ^{90}Sr, while Cases 2 and 3 are calculated using fission yields. Fission product yields used for Cases 2 and 3 are cumulative, fast-spectrum values.[3] This source does not identify ^{245}Cm as fissionable, and it is not included in the fission yield calculation. Fission of short-lived species such as ^{237}U, ^{238}Np, ^{242}Cm, etc. is neglected since these are present only in small amounts.

Decay heat output was calculated using daughter decay, representing 61 radionuclides in six decay chains, plus nine fission products. The approach neglects short-lived fission products because decay storage is included in the analysis. The results are not significantly different from the heat output from only the 29 radionuclides that were identified in the Yucca Mountain performance assessment, so the smaller set is used in this analysis (Figure 1). Daughter products do not make a relative contribution to heat output until after 10,000 yr when the total waste heat output is much less.

Some type of normalization is needed for comparing heat output and other attributes of waste streams from disparate fuel cycles such as Cases 2 and 3. For this analysis heat output is normalized to the electrical energy produced (W/GWe-d), estimated using binding energy and thermal efficiency of 32%. Normalization constants expressing electrical energy per initial metric tons heavy metal (MTHM) are shown in Table II. Using these the decay heat output characteristics for each case are readily compared (Figure 2). Cases 2 and 3 slightly increase the heat output from short-lived fission products (SLFPs) in early time, and decrease heat output from TRUs after a few hundred years.

III. REPOSITORY NEAR-FIELD TEMPERATURE

A reference thermal model is used to represent conductive heat transport in the near field of a geologic repository, assuming a simple line-loading concept similar to that proposed for Yucca Mountain. Similar

Table I. Representative Cases for Waste Thermal Evaluation for Advanced Fuel Cycles

	Implementation	Representing	Reference Comparison
Reference	Used fuel (nominal burnup 40 GWth-d/t)	Direct disposal	Current used fuel inventory
Case 1	Used fuel with SLFPs removed	Decay storage of used LWR fuel (>100 yr) and direct disposal	Similar to French system, i.e., HLW decay storage
Case 2	Used fuel radionuclide content with TRUs separated and fissioned	Reprocessing waste containing fission products and non-TRU constituents of used LWR fuel	Full recycle LWR-MOX-TRU option [7]
Case 3	Used fuel actinide content (A≥232) continuously recycled	Reprocessing waste containing fission products and no actinides lighter than ^{232}U or ^{232}Th	Fast U/TRU breeder alternative [8]

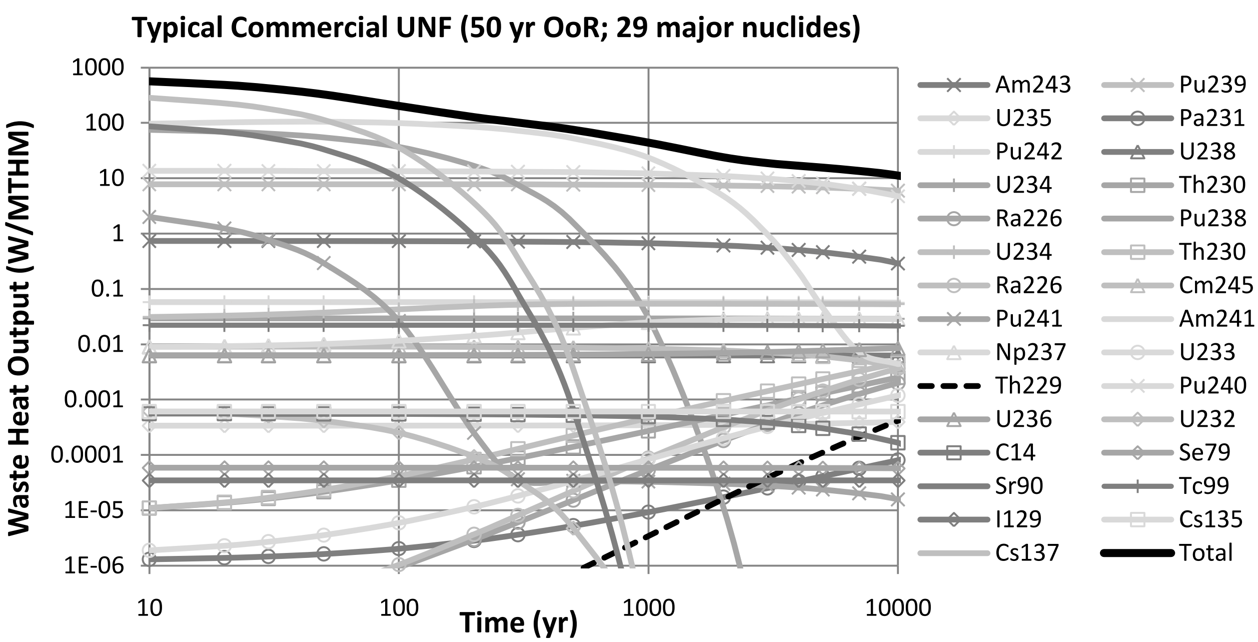

Figure 1. Waste Heat Output for 29 Major Contributing Radionuclides

Table II. Normalization Constants for Fuel Cycle Cases

	GWe-d/MTHM	Comparison Value
UNF and Case 1 – (SLFPs Removed)	15	~16 (Ref. 8, Table 1)
Case 2 (TRUs Burned)	18	(Using binding energy of 200 MeV/fission)
Case 3 (Full Recycle)	312	(Fission all actinides with mass ≥ 232)

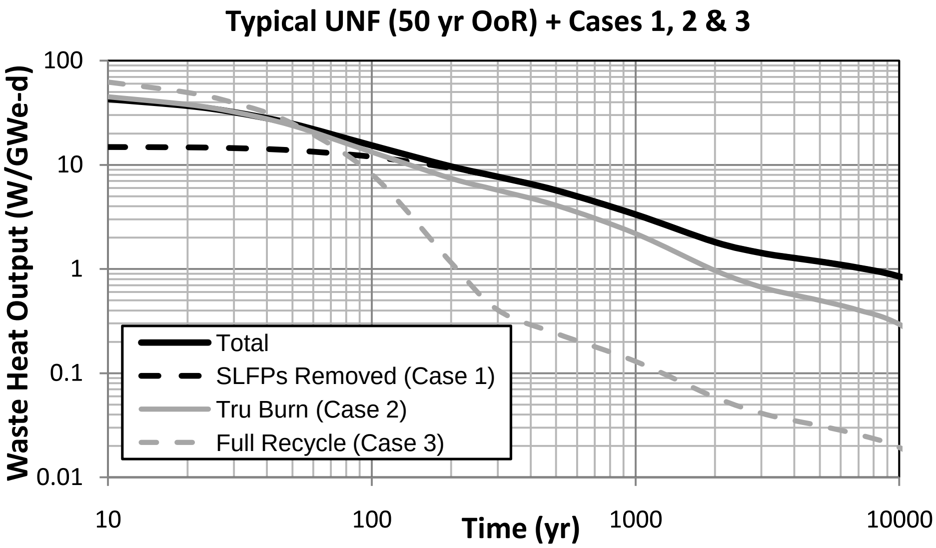

Figure 2. Normalized Waste Heat Output, Fuel Cycle Cases

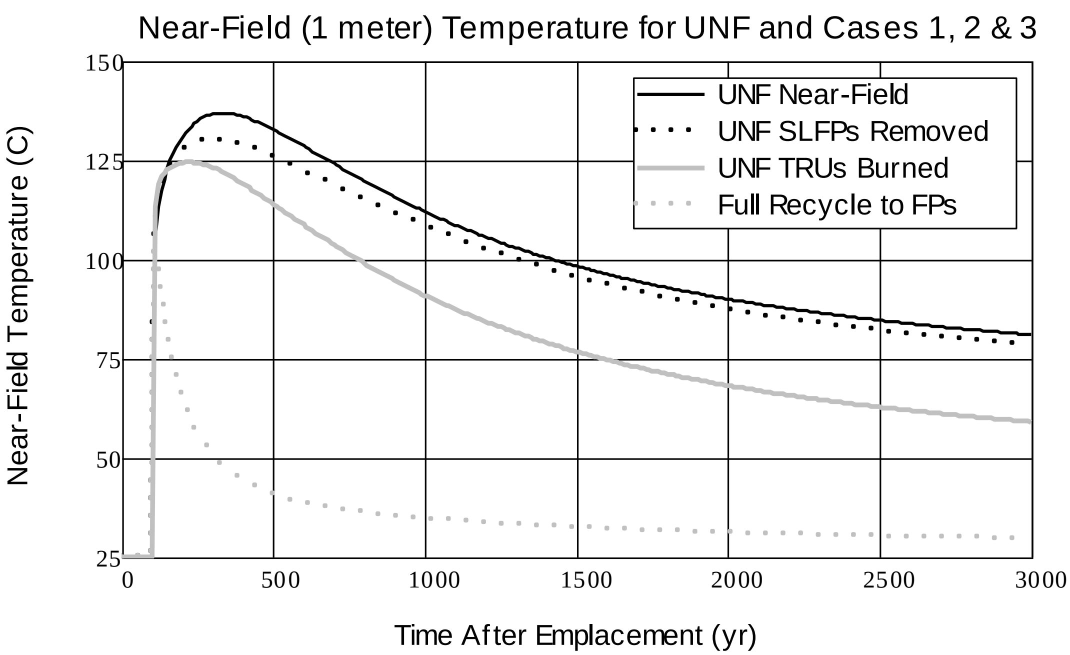

Figure 3. Near-Field Temperature for a Reference Repository Model, with the Line Loads for Fuel Cycle Cases Normalized to Electrical Energy Produced (50 yr OoR + 100 yr decay storage)

horizontal emplacement concepts have been proposed for clay[4] and granite.[5] For these calculations the line loads for the fuel cycle cases are normalized such that each waste package (5.5 meter length) corresponds to the same electrical production as for reference UNF (approximately 130 GWe-d per waste package).

Thermal calculations are done using an analytical solution approach with parallel line sources, described elsewhere.[6] The thermal reference model uses 100 yr of decay storage (or active pre-closure repository ventilation) after 2067, line-load spacing of 100 m, and host-rock thermal conductivity of 2 W/m-K (mid-range for water saturated clastic and granitic media). Near-

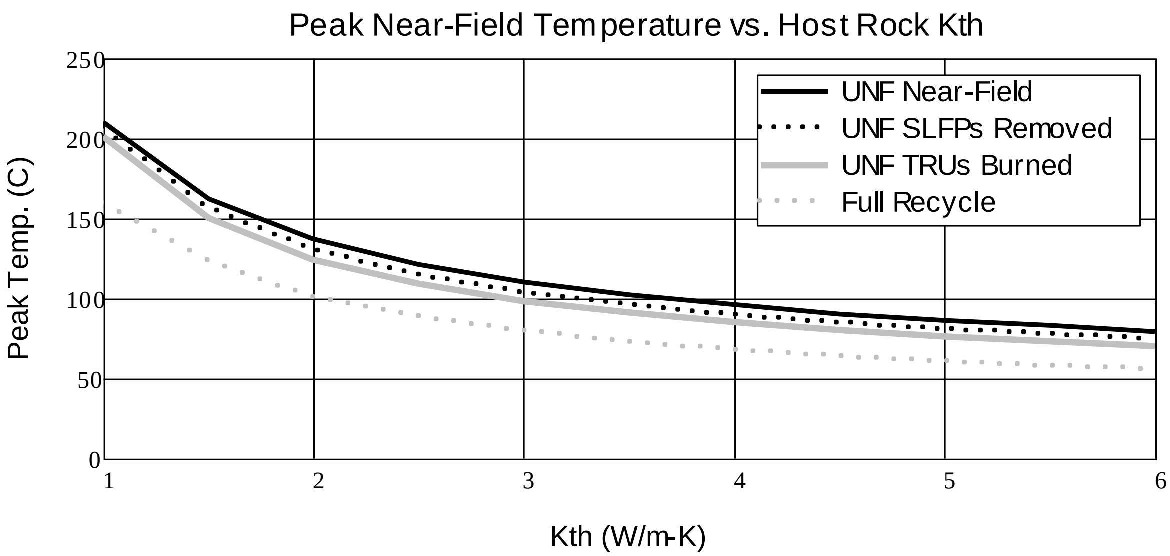

Figure 4. Peak Near-Field (1 meter) Temperatures vs. Host Rock Thermal Conductivity, for Reference Model, Fuel Cycle Cases

field temperature is calculated at a point 1 m from the centerline of a line-loaded opening (Figure 3). These thermal results (Figures 1 to 3) show that TRUs in used fuel dominate heat output after a few hundred years, and that burning the TRUs (preserving the remainder of each decay chain) only moderately decreases the duration of elevated near-field temperatures. Normalizing to electrical energy provides analternative to considering waste mass or volume, in these comparisons.

The boiling temperature for groundwater is an indicator of thermal-hydrologic-chemical-mechanical processes that complicate performance analyses. For unsaturated or shallow host media the boiling temperature may be as low as 100°C, while much higher boiling temperatures are possible for deeper, saturated host media. With “fine tuning” of thermal loading parameters (e.g., line load density and spacing, or fuel age) any of these cases (Figure 3) could be implemented so as to limit peak temperatures to 100°C or lower. Salt has the highest thrmal conductivity of any medium considered (~5 W/m-K). In the reference repository calculations for salt, given the decay storage duration and loading density (~5 W/m^2) assumed here, thermal conductivity is great enough that peak temperature never exceeds 100°C (Figure 4). This result is comparable to a previously published sensitivity case that assumed 50 years of decay storage prior to emplacement.[9] Higher peak temperatures on the order of 200°C or greater were also calculated for cases with less decay storage (<50 years), and greater loading density (>30 W/m^2).[9]

IV. RADIOTOXICITY

Fission of TRUs produces the same fission products, in similar amounts as fission of ^{235}U or ^{239}Pu, thereby increasing near-term heat generation and changing the radionuclide inventory associated with potential repository dose effects. Radiotoxicity comparisons are needed for full evaluation of the benefits from burning TRUs for thermal management or to reduce radiotoxicity. Potential radiotoxicity was calculated using the biosphere dose conversion factors for the Yucca Mountain groundwater pathway.[2] This quantity was normalized by the electrical energy produced (Table II), which has an important effect on the comparison of fission product toxicity among the fuel cycle cases. The initial toxicity of UNF was set to unity, and used to normalize all other times and cases (Figure 5).

The calculation shows that radiotoxicity from TRUs present in used fuel decreases gradually, due to Pu isotopes with half-lives greater than 10,000 years. Short-lived fission products readily decay, leaving only long-lived fission products with low activity. The “other” category of radionuclides plotted in Figure 5 includes decay series daughters (minus TRUs) for Case 2, and decay series daughters (mass number less than 232) for Case 3. The increasing activity of these “other” nuclides with time is caused by in-growth as the remaining nuclides in the series approach secular equilibrium.

Fission products are produced in approximate proportion to energy produced, for all fuel cycle cases. Short-lived fission products are not important for long-term radiotoxicity since they will fully decay to stable daughters. Long-lived fission products comprise a minor contribution to radiotoxicity, considering the entire waste inventory (Figure 5). If used fuel is fissioned to produce useable energy, while burning TRUs (Case 2) or continuously recycling actinides (Case 3), then there is no significant, relative difference in potential radiotoxicity normalized to the energy generated. Fuel cycle strategies that separate TRUs for irradiation as targets, still produce useable fission energy.

Long-lived fission products may be the only radionuclides released from a repository, or the only ones potentially capable of transiting the engineered and natural barriers, causing significant dose to members of the public in 10^6 years. Safety analyses for repositories in clay or shale[2,10] show that long-lived fission products would contribute nearly all of the potential dose in 10^6 yr (Figure 6). In the French safety assessment, only anionic species containing ^{129}I, ^{36}Cl, and ^{79}Se were transported to the biosphere at appreciable rates.[4] Hansen et al.[10] corroborated these results, and showed that ^{135}Cs and ^{237}Np may also contribute to a 10^6 year assessment, in late time, depending on chemical speciation and transport parameters. Accordingly, unless transmutation of TRUs and other actinides is associated with recovery of the fission energy produced, the dose effect (normalized to electrical energy produced) from long-lived fission products could actually increase compared to the reference case.

V. SUMMARY

Waste heat generation, repository temperature, and waste radiotoxicity were evaluated using three idealized fuel cycle cases (Table I) in addition to reference UNF. Heat output was normalized to electrical energy produced, simplifying thermal analysis of alternative fuel cycles, especially if waste mass and volume can be accommodated using various container and engineered barrier system configurations. Using a reference repository thermal model, the peak near-field temperature for these cases is shown to be in the range 100 to 130°C, indicating that any of the cases considered can be thermally “fine tuned” (line loading density, decay storage) to limit temperatures as required. Whereas transmutation of TRUs has been proposed to limit repository temperatures after decay of short-lived fission products, the repository concept of operations (drift spacing, decay storage, waste packaging, active ventilation, etc.) can be readily adjusted to accomplish the same effect.

The potential radiotoxicity from long-lived fission products, normalized to electricity produced, is effectively the same for all three fuel cycle cases. This is especially important for a repository in clay or shale, where LLFPs are the major contributors to projected dose. Thus, burning of TRUs (conversion to fission products) may decrease overall radiotoxicity, but without significantly changing the toxicity of fission products, or the projected dose for a clay/shale repository, if electrical energy is produced and taken into account (Figure 5). Separation of long-lived fission products, and direct transmutation, have limited applicability with attendant technical and economic challenges.[11] Whatever approach is taken to manage long-lived fission products, it should consider the entire system including geologic disposal, and the impacts should be normalized to the benefits, i.e., to the useable energy produced.

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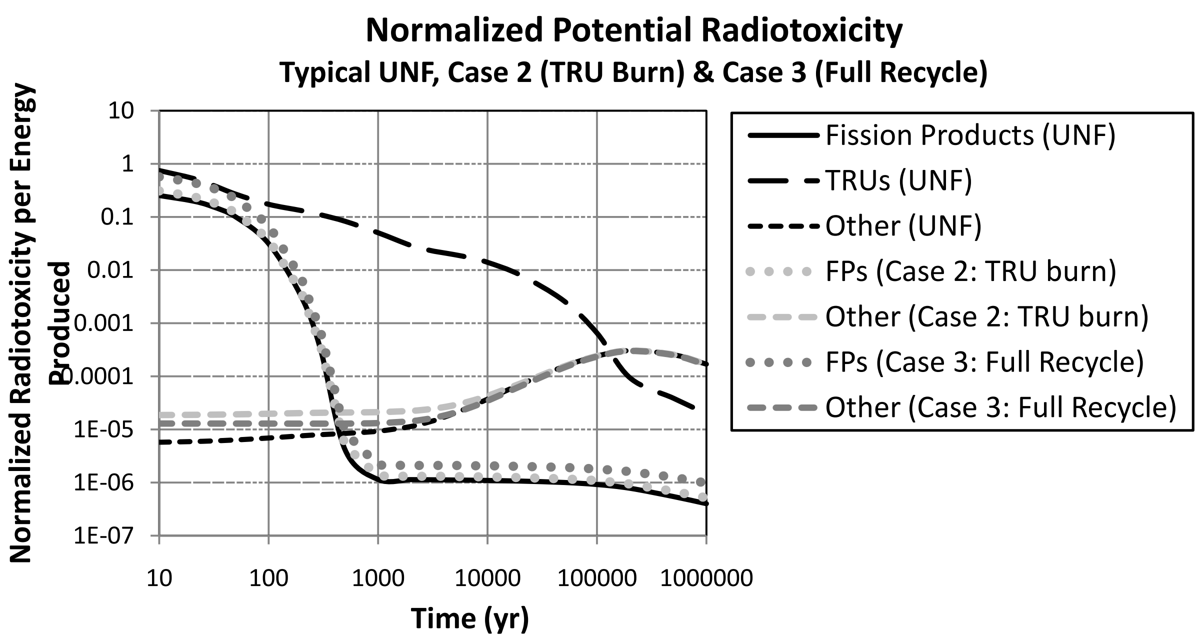

Figure 5. Normalized Potential Radiotoxicity, Fuel Cycle Cases

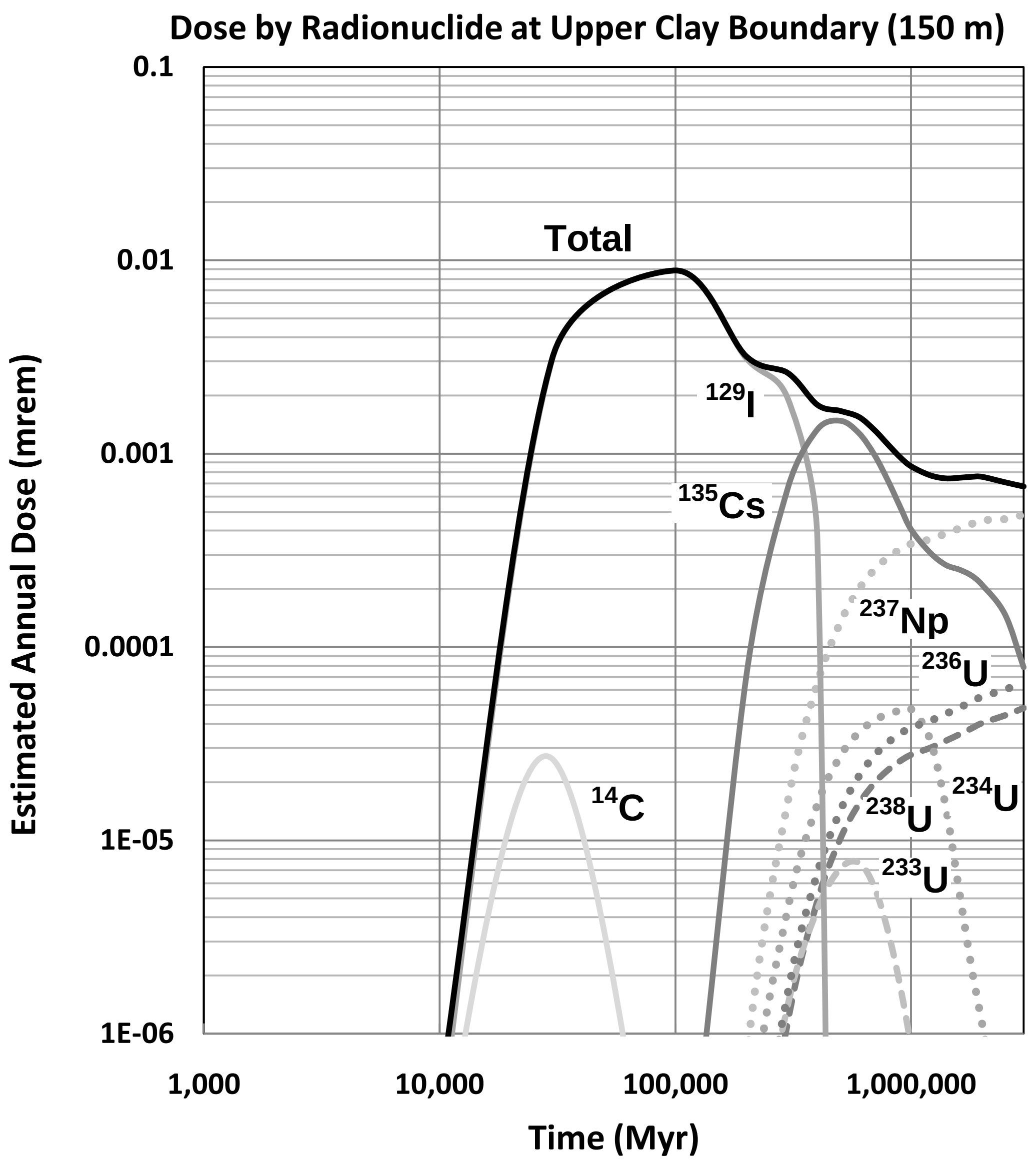

Figure 6. Performance Analysis Result for Annual Dose for a Generic Used Fuel Repository in Clay or Shale Media (from Ref. 10)