PNNL-13020

A Roadmap for Developing ATW Technology:

Performance Assessment of a Geological Repository Containing High-Level Waste and Waste from Accelerator Transmutation of Waste (ATW)

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Performance Assessment of a Geological Repository Containing High-Level Waste and Waste from Accelerator Transmutation of Waste (ATW)

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Summary

A total system performance assessment (TSPA) was conducted to determine the effects of applying accelerator transmutation of waste (ATW) to spent nuclear fuel in the proposed geologic repository at Yucca Mountain. The assessment was based on the TSPA conducted for the viability assessment (TSPA-VA) and assumes the same base case repository conditions. The base case repository contains 70,000 metric tons (t) heavy metal composed of 63,000 t of commercial spent nuclear fuel (CNSF), 2,333 t of U.S. Department of Energy spent nuclear fuel (DSNF), and 4,667 t of high-level waste. The dose history of an individual using water from a well 20 kilometers from the repository for the base case is compared to the following scenarios:

- The repository in which CSNF has undergone ATW treatment
- The repository in which both CSNF and DSNF have undergone ATW treatment
- A comparison of untreated and ATW-treated CSNF
- A comparison of untreated and ATW-treated CSNF and DSNF
- Sensitivity of dose to a factor of ten less waste packages due to ATW treatment.

The dose histories for these scenarios are plotted for 20,000 and 1,000,000 years.

For the ATW treatment, actinide elements are reduced in the spent fuel radionuclide inventory by 99.9% and long-lived fission products are reduced by 95%. The wastes arising from the ATW treatment is assumed to be disposed in Zircaloy canisters that are equivalent in long-term degradation to the cladding on the commercial spent fuel. The dissolution characteristics of wastes arising from ATW treatment are assumed to be equivalent to the high-level waste glass.

The assessment results indicate that ATW treatment of CSNF lowers the dose from the repository by as much as four orders of magnitude for the first 8,000 years. After that, the dose is lowered by about a factor of five because of the contribution from DSNF. When both CSNF and DSNF are treated, the period of lowered dose extends to11,000 years, after which the dose is lowered by only about two orders of magnitude due to the contribution of high-level waste. Comparison of treated and untreated spent fuel shows about a four-order-of-magnitude reduction in dose due to ATW treatment. The reduction in the number of waste packages by a factor of ten has no significant effect on long-term dose.

As research on ATW moves forward, it is recommended that performance assessments be refined to reflect the efficiency of ATW treatment of long-lived fission products and to evaluate more appropriate dissolution models for the ATW waste forms. In addition, the impact of long-term heat reduction on repository design due to actinide removal should be evaluated.

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1.0 Introduction

1.1 Background

The primary mechanism for transport of radionuclides from a repository to the human environment is expected to be by groundwater after the waste packages have failed. The waste is assumed to be disposed in metallic cylindrical packages emplaced on their side in drifts excavated in the rock of the unsaturated zone. For radionuclides to be released, the waste container and other engineered barriers must fail, allowing formation of a water film on the waste form through which release can occur by diffusion, or allowing advection (flow) through the failed package. Radionuclides leached from commercial spent nuclear fuel (CSNF), defense spent nuclear fuel (DSNF), and high-level waste (HLW) are expected to migrate out of a failed barrier system, by diffusion or advection, to the water table through the unsaturated zone and to the human environment through the saturated zone.

Flux through the unsaturated repository horizon arises from infiltration of precipitation at the ground surface minus the amount returned to the atmosphere through evaporation and transpiration. The small portion of the infiltration that remains percolates downward to the water table (top of the saturated zone) controlled by capillary and gravity forces. Heat from radioactive decay of the waste influences this downward percolation and must be considered in a performance assessment. The amount of heat present depends on the design of the repository (i.e., the spacing of repository drifts, the spacing and thermal loading of the waste packages, the type of waste, and the age of the waste). The migration of radionuclides through the unsaturated zone is a function of the percolation flux, dispersion, and retardation of radionuclides by sorption to mineral surfaces along the flow path, or by diffusion into slower percolating zones (diffusion from the fractures into the rock matrix where flow is slower). Sorption is a function of the chemistry of the specific radionuclide, the chemistry of the water, and the chemistry of the minerals along the flow path. Dispersion is the spreading of the plume caused by the tortuous paths that the radionuclides follow through openings (pores and fractures) in the rock.

At the water table, the concentration of radionuclides is diluted by the flow in the saturated zone. The radionuclides are then transported through the saturated zone to a defined point where their concentration can be compared to regulatory standards. Along the transport pathway, the concentration of radionuclides is reduced by dispersion (spreading of the plume) and delayed by retardation (lagging behind the flow due to sorption and matrix diffusion). The delay caused by retardation may allow concentrations to be reduced through radioactive decay, depending on the delay time in relation to the half-life of the specific radionuclide.

In the human environment, the concentration of radionuclides over time is used to calculate dose to an individual based on a scenario that the individual is sustained by using groundwater from a well for production of all food, for household use, and for drinking water. The peak dose from a specific radionuclide occurs at the time of peak concentration of that radionuclide in the groundwater. Peak concentration is controlled by all of the processes discussed above (i.e., peaks for different chemical species occur at different times due to differences in solubility and sorption along the transport path). The new standard for Yucca Mountain is expected to be dose-based, and therefore the analyses in the Total

System Performance Assessment-Viability Assessment (TSPA-VA) follows U.S. Department of Energy (DOE) interim guidance and reports dose from all sources to an individual located 20 kilometers down the hydraulic gradient from the repository (DOE 1998). A schematic diagram of the transport pathway from the repository to humans is shown in Figure 1.

The base case repository for the TSPA-VA contains 70,000 metric tons heavy metal (MTHM) of radioactive waste. The wastes are composed of 63,000 MTHM of CSNF, 4,667 MTHM of HLW, and 2,333 MTHM of DSNF. The area mass loading of the repository is 85 metric tons uranium per acre based on CSNF (about 75kW/acre). The wastes are assumed to be contained in packages that have a corrosion allowance outer barrier of carbon steel and a corrosion resistant inner barrier of Alloy 22 (a nickel alloy). The first package fails at about 4,000 years and about 6.2% of the packages have failed by 100,000 years.

Because of the long duration of repository analyses, the climate is expected to change. For the first 5,000 years the current dry climate is assumed to exist (unsaturated-zone flux of 7.7 mm/yr), and periods of dry climate are expected to last 10,000 years thereafter. The first dry climate is followed by a period of long-term average climate (unsaturated-zone flux of 42 mm/yr) that lasts for 90,000 years. Thereafter, the dry climates alternate with periods of long-term average climate. In addition, a super pluvial climate (unsaturated-zone flux of 110 mm/yr) is assumed to occur at about 250,000 years and 750,000 years with durations of about 10,000 years. The saturated-zone fluxes that correspond to the three climates (current dry, long-term average, and super pluvial) are 0.6 m/yr, 2.3 m/yr, and 3.7 m/yr, respectively.

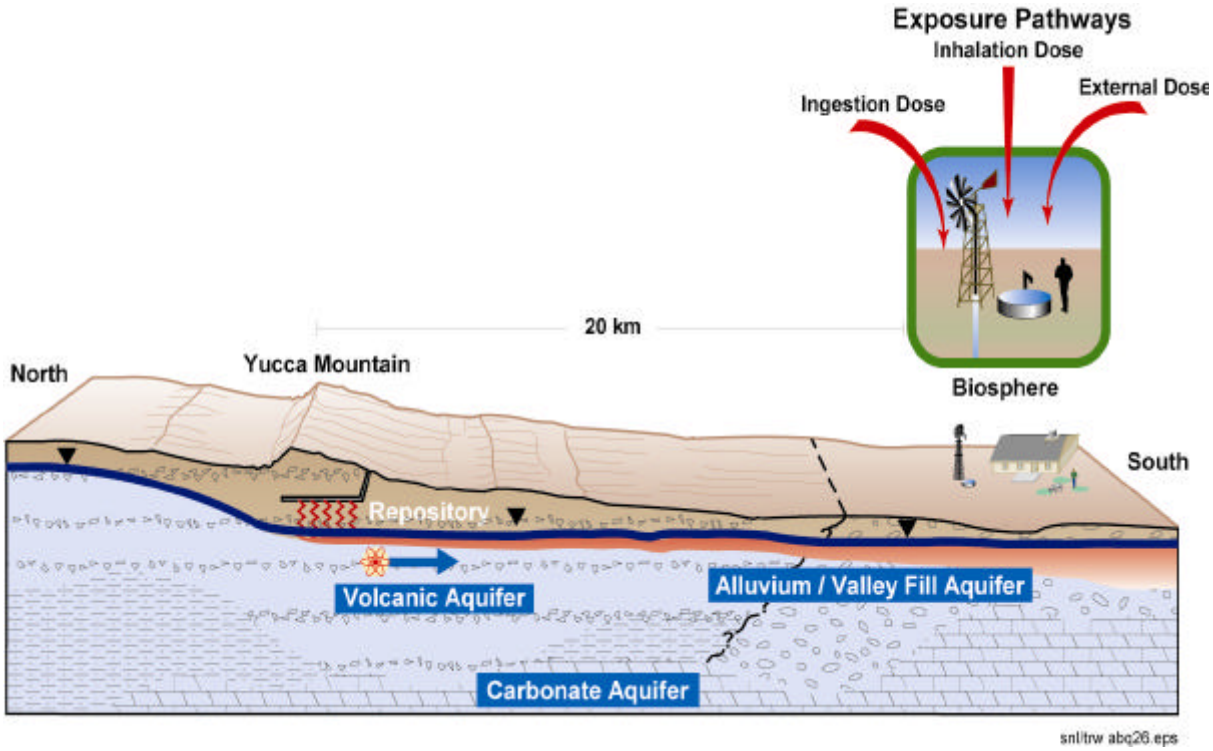

Figure 1. Schematic Diagram of the Transport Pathway from the Repository to the Human Environment.

The biosphere model considers dose from all sources to an individual using water from a well 20 kilometers down the hydraulic gradient from the repository. The dose conversion factors are based on lifestyles of current residents of the Amargosa Valley region, and the dose pathways include ingestion, inhalation, and external dose.

The performance assessment for the waste resulting from Accelerator Transmutation of Waste (ATW) is based on the model used for the TSPA-VA, briefly described above. The analyses for the TSPA-VA are summarized in volume three of the Viability Assessment of a Repository at Yucca Mountain (DOE 1998). A more detailed discussion of the TSPA-VA analyses is contained in the Technical Basis Document for TSPA-VA (CRWMS M&O, Chapters 1-11).

1.2 Purpose

The purpose of assessing the dose from ATW treatment of CSNF and DSNF is to guide the work of the ATW Separations Technologies and Waste Forms Technical Working Group. The results are based on the TSPA-VA model, which will change significantly as the repository design is optimized for the license application for the repository (CRWMS M&O 1999). For the license application repository design, doses are expected to be considerably lower than for the TSPA-VA base case used for these analyses. However, the same relative difference between the base case results and the waste from ATW would remain using the license application model (i.e., both the base case and ATW scenarios would be changed by the same amount).

1.3 Scope

The expected-value dose history at 20 kilometers for the TSPA-VA base case is compared to a repository containing waste from application of ATW to CSNF and a repository containing ATW-treated CSNF and DSNF. Comparisons are made between treated and untreated CSNF (no HLW or DSNF) and between treated and untreated CSNF and DSNF (no HLW). Sensitivity cases are presented for a factor of ten fewer waste packages for CSNF and DSNF. Analyses for both 20,000 years and 1,000,000 years are presented.

2.0 Approach and Major Assumptions

2.1 Approach

The performance assessment of wastes from ATW is based on placing the wastes in the environment of the repository analyzed for the TSPA-VA (DOE 1998) and calculating the expected-value dose they produce. The results from a repository containing ATW-treated wastes are compared to the repository without ATW treatment (e.g., the base case TSPA-VA repository).

2.2 Assumptions

The ATW treatment is assumed to reduce the actinide elements in the CSNF and DSNF by 99.9%. The long-lived fission products, 129I and 99Tc, are assumed to be reduced by 95%. These reductions are based on the separations technology projected reductions (Laidler et al. 1999). They assume complete transmutation of the 129I and 99Tc or that the remaining 129I and 99Tc is packaged so that it does not affect the calculated dose. The amount of 129I and 99Tc remaining in the waste was increased to reflect the amount of these fission products generated by ATW. This assumption may not be conservative for 129I and 99Tc because the efficiency of transmuting these long-lived fission products is likely to be lower than assumed.

The waste from ATW treatment is assumed to be disposed in the TSPA-VA base case repository. The repository is assumed to be the same size, have the same thermal characteristics, and contain the same number of waste packages as the TSPA-VA base case repository. Because the number of waste packages might be reduced in the future, a sensitivity case is analyzed with a factor of ten fewer waste packages.

The assumption is made that these waste forms that arise from ATW have dissolution rates at least as good as HLW glass, and the HLW dissolution model is used. This is likely to be a conservative assumption (i.e., the dose calculated is higher than it would be if the dissolution model for the ATW waste forms was used). The waste form from the initial separation is glass-bonded sodalite, and the waste form from separation of ATW fuel is a zirconium-steel alloy (Laidler et al. 1999). The dissolution rate from these materials could be lower than that of the HLW glass.

The CSNF has Zircaloy cladding, which is a barrier to radionuclide release that reduces the dose by about three orders of magnitude at times before about 10,000 years. During ATW, the Zircaloy cladding is removed. For this reason, the ATW-treated waste is assumed to be contained in a Zircaloy canister that has the same performance as the CSNF cladding. If this assumption were not made, the performance of clad CSNF and ATW-treated CSNF would be about the same (except for waste form dissolution differences). The three order of magnitude increase caused by cladding removal would be canceled by the similar reduction in radionuclide inventory.

3.0 Discussion Of Results

To compare the following results, the expected-value dose history at 20 kilometers over 100,000 years from the TSPA-VA base case repository is shown in Figure 2. Figure 2 shows that the dose from the repository is primarily from three radionuclides (99Tc, 129I, and 237Np). The dose from the base case repository is from 99Tc and 129I for about the first 50,000 years, after which the dose is primarily from 237Np (Figure 2).

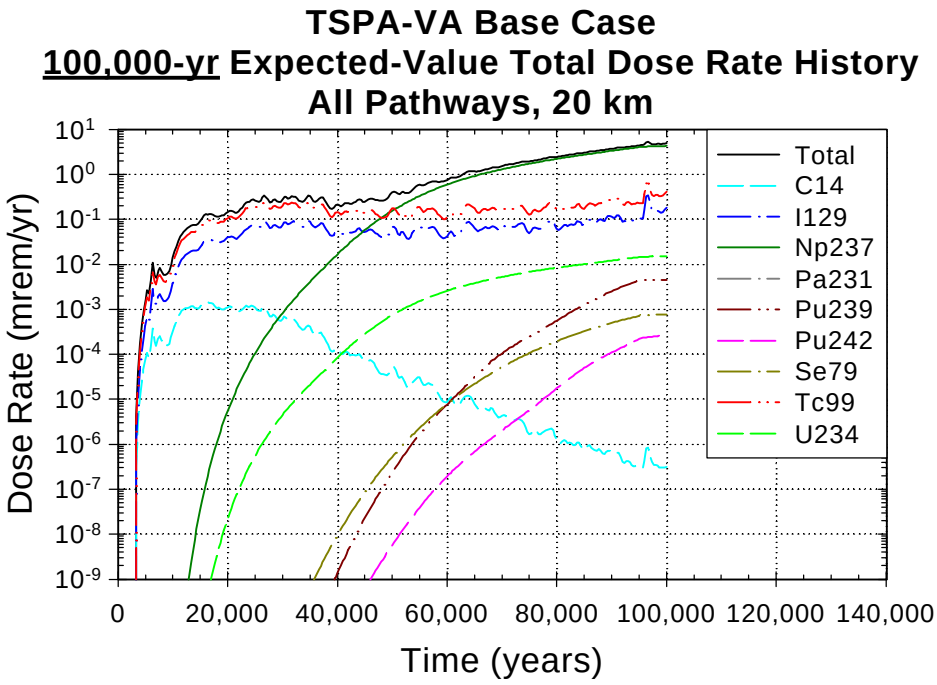

Figure 2. Expected-Value Dose History Over 100,000 Years to an Individual using Water from a Well 20 Kilometers from the Repository Analyzed in the Total System Performance Assessment-Viability Assessment Base Case.

The results from the TSPA-VA base case repository and a repository in which CSNF has been treated by ATW are shown in Figure 3. The dose from the treated CSNF is about four orders of magnitude lower than the base case out to about 8,000 years, where it increases dramatically because of the contribution from DSNF. The doses from the two repositories (base case and ATW treatment of CSNF) differ by about a factor of five at 20,000 years because of the contribution of DSNF (Figure 3). When the DSNF is also treated, the doses differ by about four orders of magnitude to about 11,000 years, where there is a rapid increase in dose from the HLW (Figure 4).

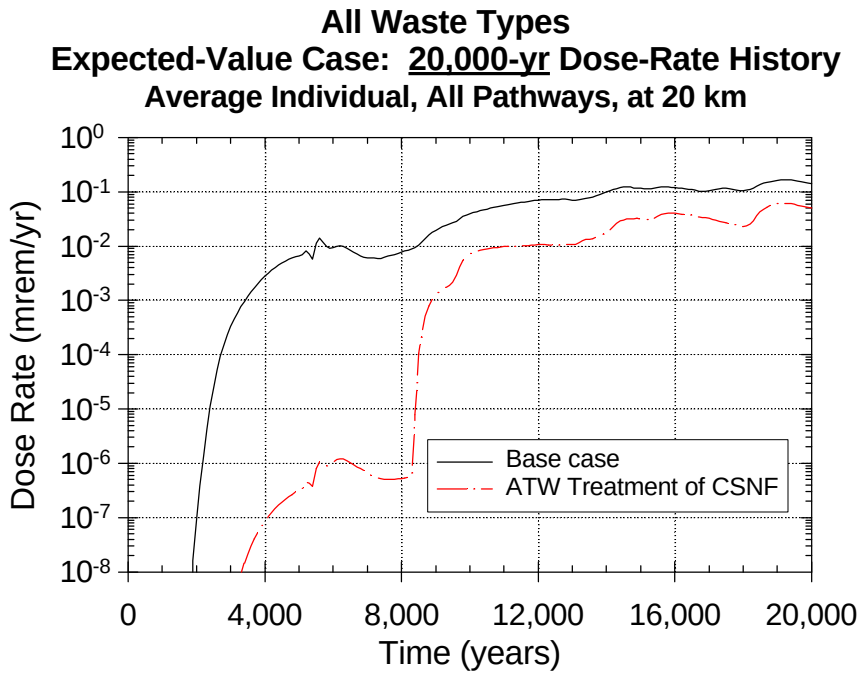

Figure 3. Expected-Value Dose History at 20 Kilometers Over 20,000 Years from the Base-Case Repository Compared to a Repository Where the CSNF has Undergone ATW Treatment.

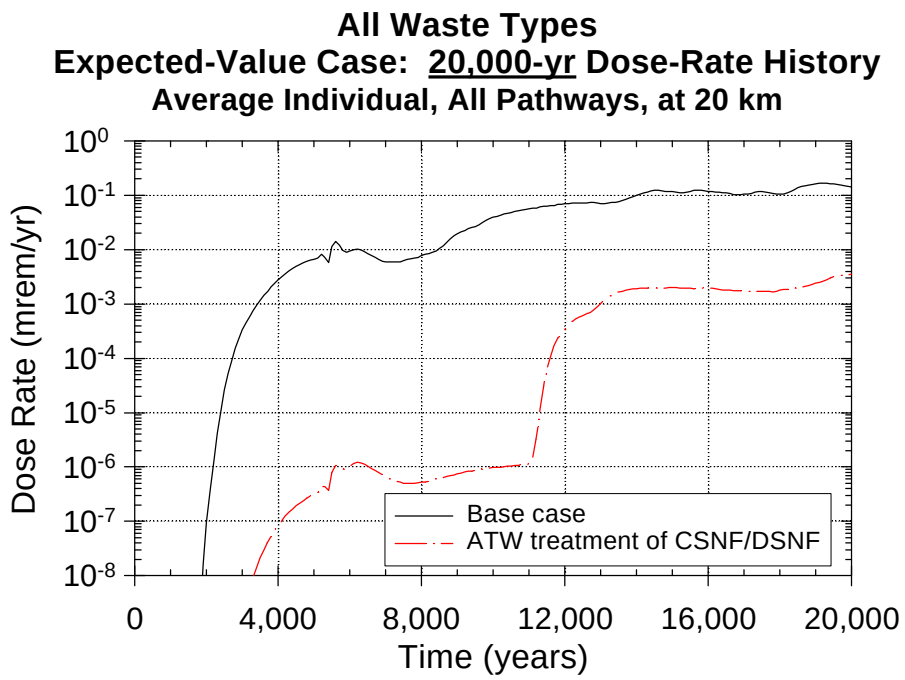

Figure 4. Expected-Value Dose History at 20 Kilometers Over 20,000 Years from the Base-Case Repository Compared to a Repository Where the CSNF and DSNF have Undergone ATW Treatment.

Beyond about 12,000 years, the doses differ by about two orders of magnitude out to 20,000 years. When the untreated CSNF is compared to treated CSNF, the four order of magnitude difference remains for the entire 20,000-year period (Figure 5). About three of the four orders of magnitude differences are due to the reduction in radionuclide inventory in the waste, and one order of magnitude difference is due to the change in dissolution model. For the untreated CSNF the commercial spent fuel dissolution model is used, and for the treated fuel the HLW glass dissolution model is used.

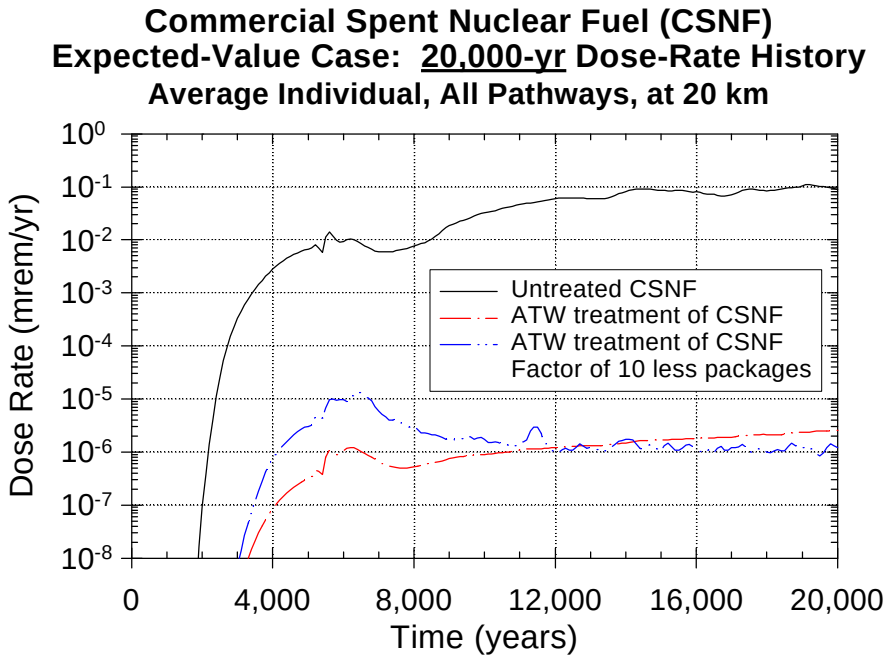

Figure 5. Expected-Value Dose History at 20 Kilometers Over 20,000 Years from Untreated CSNF, ATW-Treated CSNF, and ATW-Treated CSNF Contained in a Factor of Ten Fewer Packages.

Figure 5 also shows the change in dose when the number of waste packages is reduced by a factor of ten. This reduction causes the dose peak from long-lived fission products to increase because there is more inventory of 99Tc and 129I in each package. These radionuclides are released rapidly from the failed packages because of their high solubility. After 12,000 years, the decrease in the number of packages causes the dose to decrease somewhat because there are fewer failed packages releasing 237Np at its solubility limit.

Figure 6 is similar to Figure 5 except both CSNF and DSNF are shown. Here it is interesting to note that the dose curve for untreated CSNF and DSNF is the same as for the base case repository (compare the upper curve of Figure 6 to the base case of Figure 4). The contribution from DSNF to the dose from the base case repository can be observed by comparing the upper curves of Figures 5 and 6. Here the dose from CSNF is somewhat lower than that from CSNF and DSNF after about 11,000 years. The discussion above relating to the reduction in the number of waste packages also applies to the results shown on Figure 6.

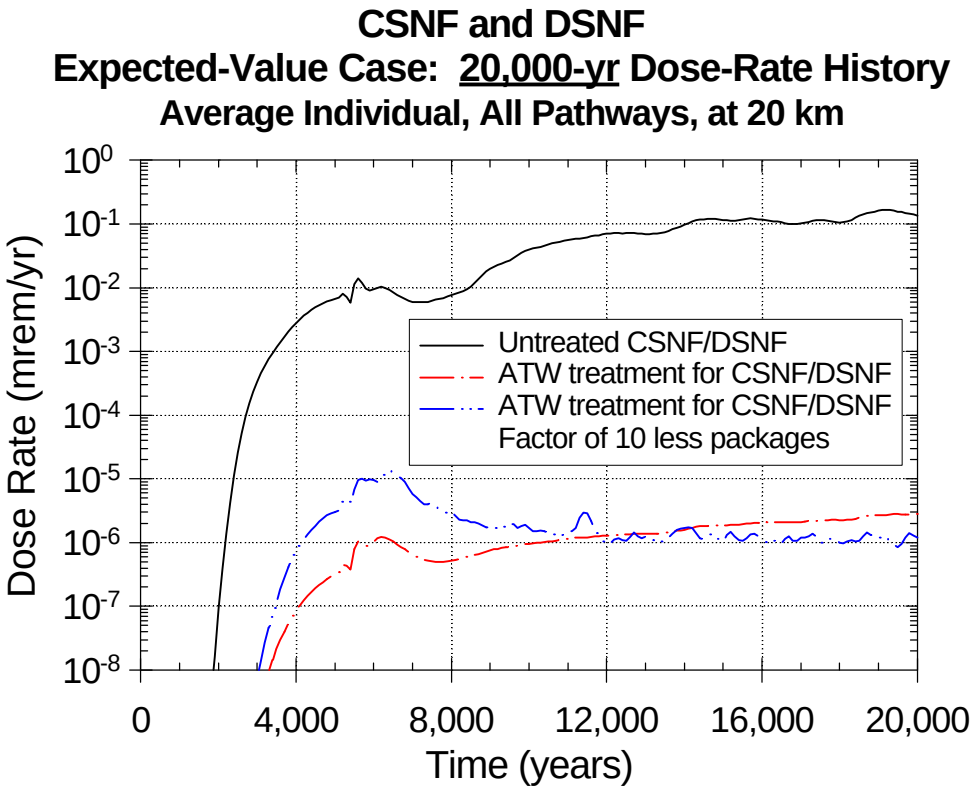

Figure 6. Expected-Value Dose History at 20 Kilometers Over 20,000 Years from Untreated CSNF and DSNF, ATW-Treated CSNF and DSNF, and ATW-Treated CSNF and DSNF Contained in a Factor of Ten Fewer Packages.

Over a one-million-year period, the peak dose is reduced by about an order of magnitude by ATW treatment of CSNF (Figure 7). When both CSNF and DSNF are treated, the peak dose is reduced by about two orders of magnitude (Figure 8). The dose peak under the ATW treatment scenarios shown in Figure 7 and Figure 8 is caused by the contribution from DSNF and HLW, respectively. Figure 9 shows a comparison of treated and untreated CSNF to show the full affect of ATW treatment. Here, the four order of magnitude difference is caused by a combination of reduction in radionuclide inventory (about three orders of magnitude) and a change in dissolution model (about one order of magnitude). Also, there is little difference in the peak dose from a factor of ten fewer waste packages (Figures 9 and 10). Figure 10 shows a comparison of treated and untreated DSNF and CSNF. There is little difference in peak dose between the scenario of treating both DSNF and CSNF (Figure 10) and treating only CSNF (Figure 9). This small difference is because DSNF contributes only a few percent of the total dose from the base case repository. In addition, the ATW treated DSNF is in Zircaloy canisters while in the base case it is assumed to be unclad.

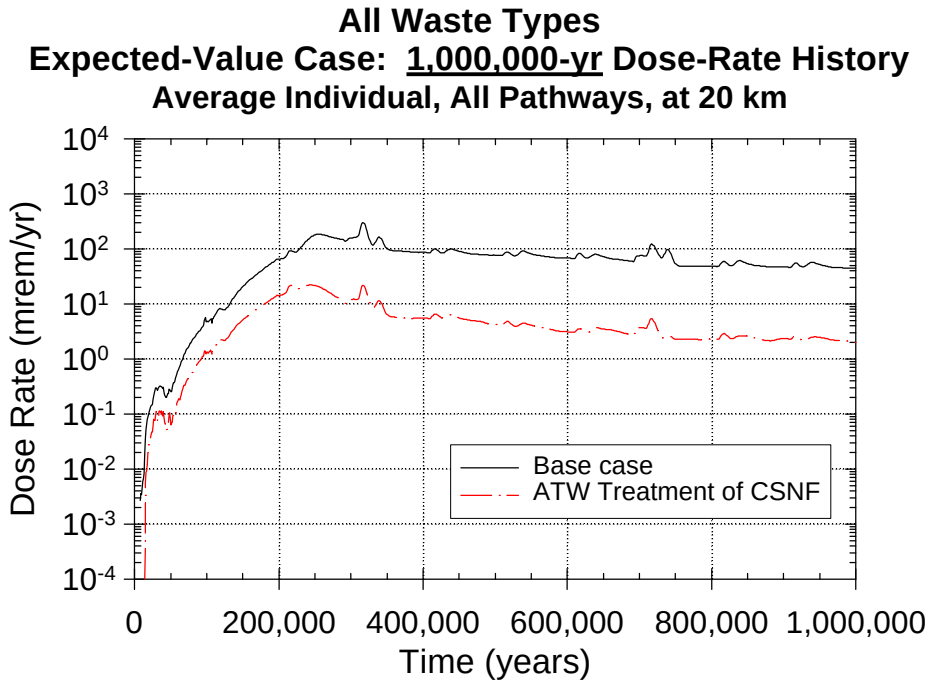

Figure 7. Expected-Value Dose History at 20 Kilometers Over 1,000,000 Years from the Base-Case Repository Compared to a Repository where the CSNF has Undergone ATW Treatment.

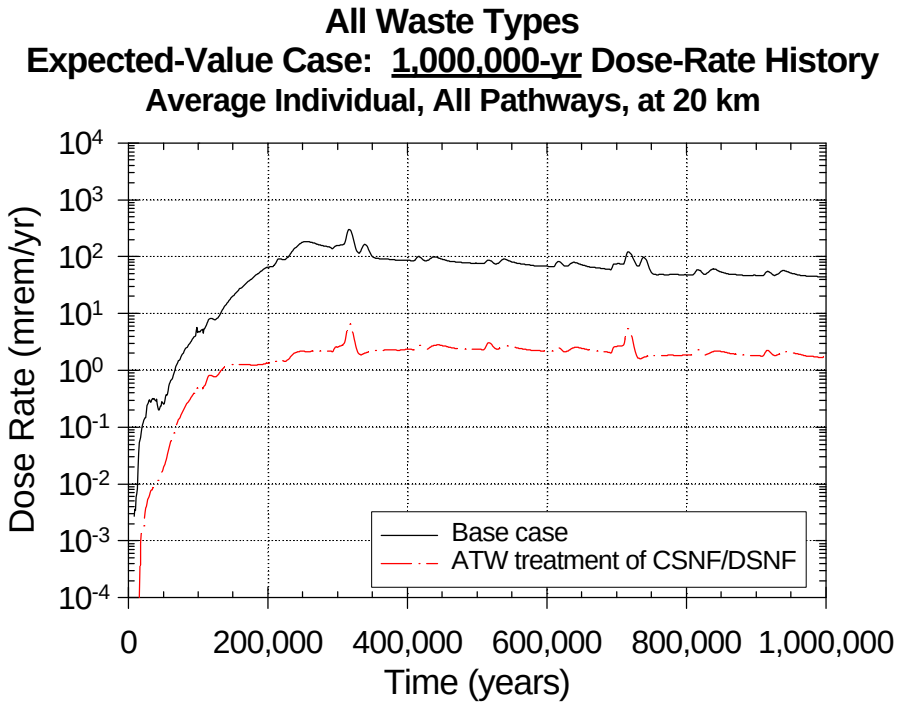

Figure 8. Expected-Value Dose History at 20 Kilometers Over 1,000,000 years from the Base-Case Compared to a Repository where the CSNF and DSNF have Undergone ATW Treatment.

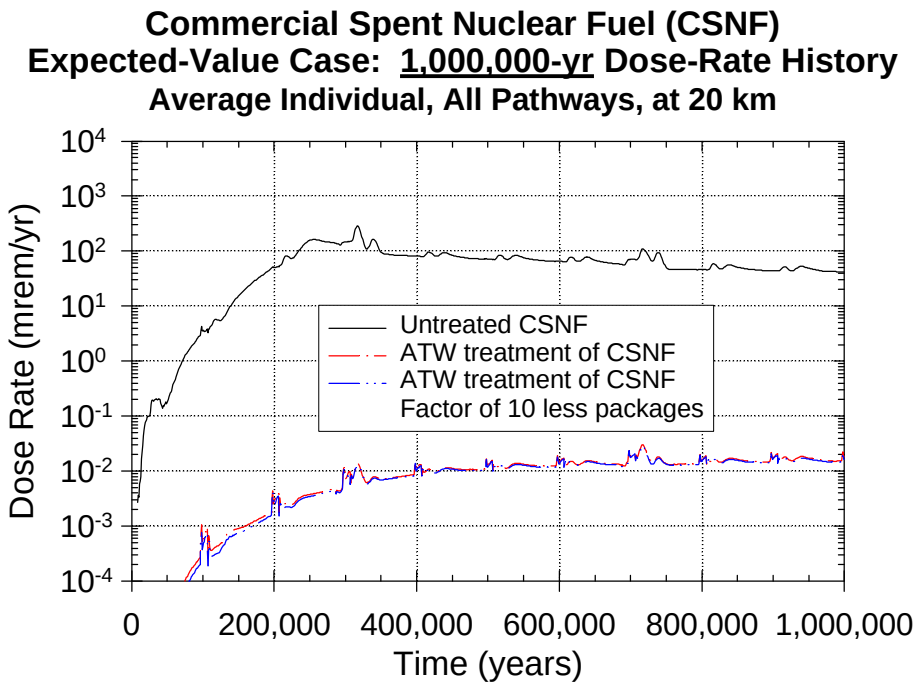

Figure 9. Expected-Value Dose History at 20 Kilometers Over 1,000,000 Years from Untreated CSNF, ATW-Treated CSNF, and ATW-Treated CSNF Contained in a Factor of Ten Fewer Packages.

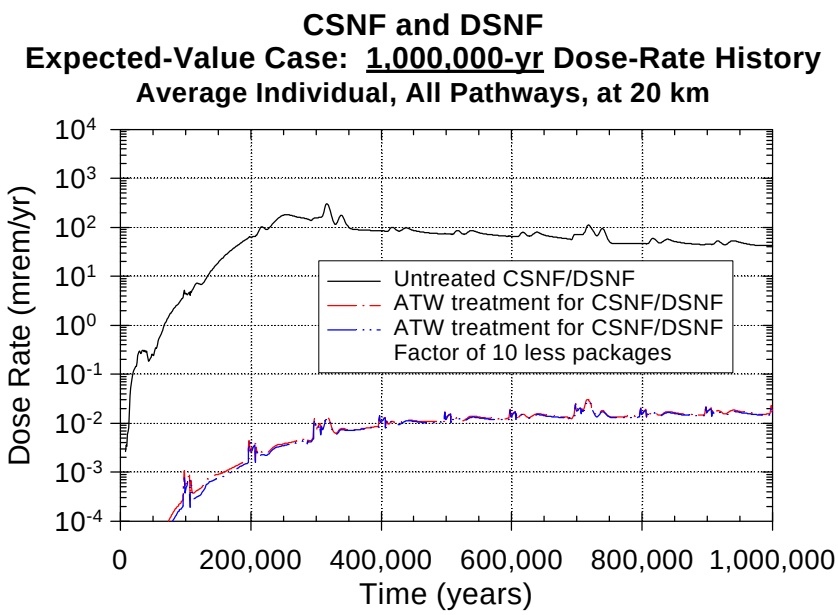

Figure 10. Expected-Value Dose History at 20 Kilometers Over 1,000,000 Years from Untreated CSNF and DSNF, ATW-Treated CSNF and DSNF, and ATW-Treated CSNF and DSNF Contained in a Factor of Ten Fewer Packages.

4.0 Conclusions and Recommendations

4.1 Conclusions

Based on the analyses of applying ATW to repository wastes, the following conclusions are made:

- Application of ATW to CSNF reduces the dose from the repository by four orders of magnitude out to about 8,000 years. After 8,000 years, the dose from the repository is from DSNF and is only about a factor of five lower than the base case repository at 20,000 years.

- Application of ATW to CSNF and DSNF reduces the dose from the repository by four orders of magnitude out to about 11,000 years. After 11,000 years, the dose from the repository is from HLW and is about two orders of magnitude lower than the base case repository.

- Application of ATW to CSNF lowers the peak dose from the repository by only about one order of magnitude because of the contribution from the DSNF.

- Application of ATW to both CSNF and DSNF lowers the peak dose from the repository by only about two orders of magnitude because of the contribution from HLW.

- A factor of ten reduction in the number of waste packages from application of ATW has little affect on peak dose.

4.2 Recommendations

For future work on ATW, the following recommendations for future performance assessments are made:

- Even though the same relative difference will remain between wastes from ATW and untreated wastes, the performance assessment should be updated to the repository design used for the license application.

- Changes in the repository size and thermal loading arising from ATW treatment of waste should be evaluated. The application of ATW to repository wastes could lead to changes in the repository design.

- A more appropriate dissolution model than HLW glass should be developed and used for the glass-bonded sodalite and the zirconium-steel alloy. The dissolution model for ATW waste forms would be expected to have a lower dissolution rate than HLW glass.

- Performance assessment is a useful tool in guiding trade studies in the development of ATW. Additional work is needed for the evaluation of 129I and 99Tc as the efficiency of ATW in transmuting these long-lived fission products is developed.

- Future performance assessment work for development of ATW is estimated to require approximately one-person year per year.

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Appendix A

Acronyms

Appendix A

Acronyms

ATW	Accelerator transmutation of waste
CRWMS	Civilian radioactive waste management system
CSNF	Commercial spent nuclear fuel
DOE	U.S. Department of Energy
DSNF	Defense spent nuclear fuel
HLW	High-level waste
129I	Iodine-129, an isotope of iodine
MTHM	Metric tons heavy metal
M&O	Management and operating contractor
237Np	Neptunium-237, an isotope of neptunium
99Tc	Technetium-99, an isotope of technetium
TSPA	Total system performance assessment
VA	Viability assessment