Application of Analytical Heat Transfer Models of Multi-layered Natural and Engineered Barriers in Potential High-Level Nuclear Waste Repositories - 12435

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ABSTRACT

A combination of transient heat transfer analytical solutions for a finite line source, a series of point sources, and a series of parallel infinite line sources were combined with a quasi-steady-state multi-layered cylindrical solution to simulate the temperature response of a deep geologic radioactive waste repository with multi-layered natural and engineered barriers.

This evaluation was performed to provide information to scientists and decision makers to compare candidate geologic media for a repository (crystalline rock [granite], clay, salt, and deep borehole), and to provide input for the future evaluation of the trade-off between pre-emplacement surface storage time, waste package size, and repository footprint.

This approach was selected in favor of the finite element solution typically used to analyze the temperature response because it allowed rapid comparison of a large number of alternative disposal options and design configurations. More than 100 combinations of waste form, geologic environment, repository design configuration, and surface storage times were analyzed and compared.

INTRODUCTION

The Used Fuel Disposition (UFD) Campaign within the Department of Energy's Office of Nuclear Energy (DOE-NE) Fuel Cycle Technology (FCT) program has been tasked with investigating the disposal of the nation's high-level nuclear waste (HLW) and spent nuclear fuel (SNF) from commercial nuclear power plants, for a range of potential waste forms and geologic environments.

For each waste form and geologic environment combination, there are multiple options for a repository conceptual design. Comparison of alternative scenarios using finite-element computer codes can make parametric sensitivity studies expensive and time consuming. An alternative approach uses two analytical heat transfer solutions. A transient "outside" model was developed assuming a homogeneous infinite medium to portray the temperature transient in the host rock, and a quasi-steady-state multi-layer cylindrical "inside" model was developed to represent the thermal response of the Engineered Barriers System (EBS). The "outside" model calculates a temperature transient, given decay heat data from the waste form, at the borehole or tunnel wall of a geologic repository by assuming the uniform infinite medium extends both inside and outside the "calculation radius". The "inside" model uses the temperature calculated by the "outside" model at the host rock wall surface in conjunction with the transient heat source, and calculates the thermal gradient through the EBS using a steady-state multi-layer cylindrical model solution. This approach is reasonable because the thermal mass of the EBS components is much smaller than the infinite mass of host rock surrounding the EBS. There is a short (on the order of weeks or months) transient in the EBS components when the waste is initially placed in the repository. After that the component temperatures follow the continuing temperature transient in the surrounding host rock, which slowly evolves because of its large thermal mass.

These models allowed thermal performance comparisons to address alternative disposal concepts including:

- Four host rock environments – crystalline rock (granite), clay, salt, and deep borehole
- The waste inventory considered three potential nuclear fuel cycle concepts
  - Once-through high-burnup (60 GWd/MTU) uranium oxide (UOX) light water reactor (LWR) fuels
  - Modified open cycle including mixed oxide fuels (MOX) and high-level waste glass (Co-Extraction) represents an example of thermally hotter SNF waste forms
  - A full recycle concept based on an advanced burner reactor (ABR) with three types of high-level waste (New Extraction HLW glass, electro-chemical ceramic HLW, and electro-chemical metal HLW)
- Two EBS design concepts in each media, one for used nuclear fuel assemblies (SNF), and one for HLW canisters
- Four surface storage times – 10, 50, 100, and 200 years
- Five SNF waste package assembly capacities – 1, 2, 3, 4, and 12 assemblies (evaluated for the once-through and modified-open fuel cycles)

While "co-extraction" process is similar in function to the industrial Co-Extraction™ (COEX) process deployed by AREVA, the two assume different processing methods and steps, so the product and waste streams cannot be directly compared. Similarly, the "new extraction" process and the NUEX industrial process proposed by Energy Solutions also cannot be directly compared.

Repository Design Concepts

An expert, multidisciplinary panel, consisting of representatives from four National Laboratories and DOE/NE, with experience in US and international repository programs selected basic input data to be analyzed for:

- Thermal load from a variety of waste streams,
- Repository configurations, and
- Material properties and thermal constraints

The repository configurations selected were based on current international design concepts for both SNF and HLW deep geologic disposal systems in each of the host rock types. Unlike repository designs with large open tunnels and pre-closure ventilation, all of the disposal concepts selected for this study use enclosed emplacement modes, where the waste packages are in direct contact with encapsulating engineered or natural materials. The concepts of operation for the various alternatives are discussed in [1], and in a companion paper presented at this conference [2]. Example design concepts were considered for the various media for:

- Granite (crystalline rock) – from Sweden, Finland and Japan
- Clay – from Belgium, France and Switzerland
- Salt - from Germany and the US
- Deep Borehole – from Sweden and the US

The specific design concepts in each medium type are discussed in [1]. The international design concepts were not taken verbatim, but were modified by the panel in a number of cases, specifically with respect to layout assumptions, and EBS component dimensions.

To model the wide variety of international design concepts and terminology used, a standardized EBS geometry was defined, such that each of the concepts modeled would use a subset of the generic layers of concentric EBS material layers shown in Figure 1. The specific design concepts and dimensions for the EBS components for each of the four environments, for both SNF and HLW design concepts, are given in [3] and [4].

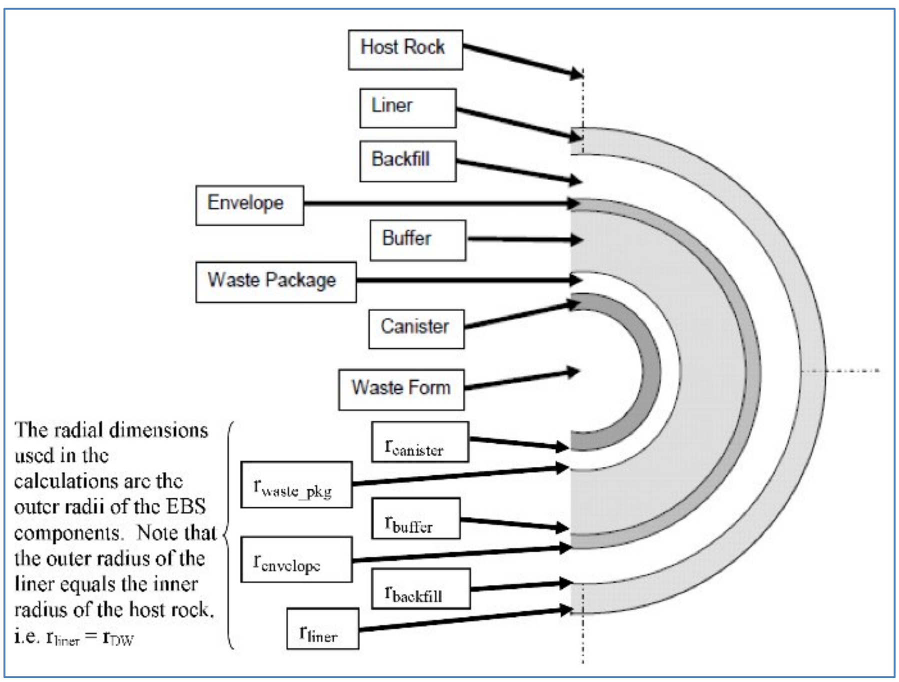

Figure 1 - Generic EBS Layer Definitions

The waste inventory and decay heat transient data per metric ton of uranium (MTU) for three potential nuclear fuel cycles were developed by SRNL [5] and [6]. Using their data, LLNL developed decay heat input curves on a per-assembly and per HLW canister basis, as a function of time-out-of-reactor, [4] which is shown in Figure 2. Note that 5-years out-of-reactor is the minimum at-reactor storage time, and would be equivalent to direct disposal with no other interim surface storage time required.

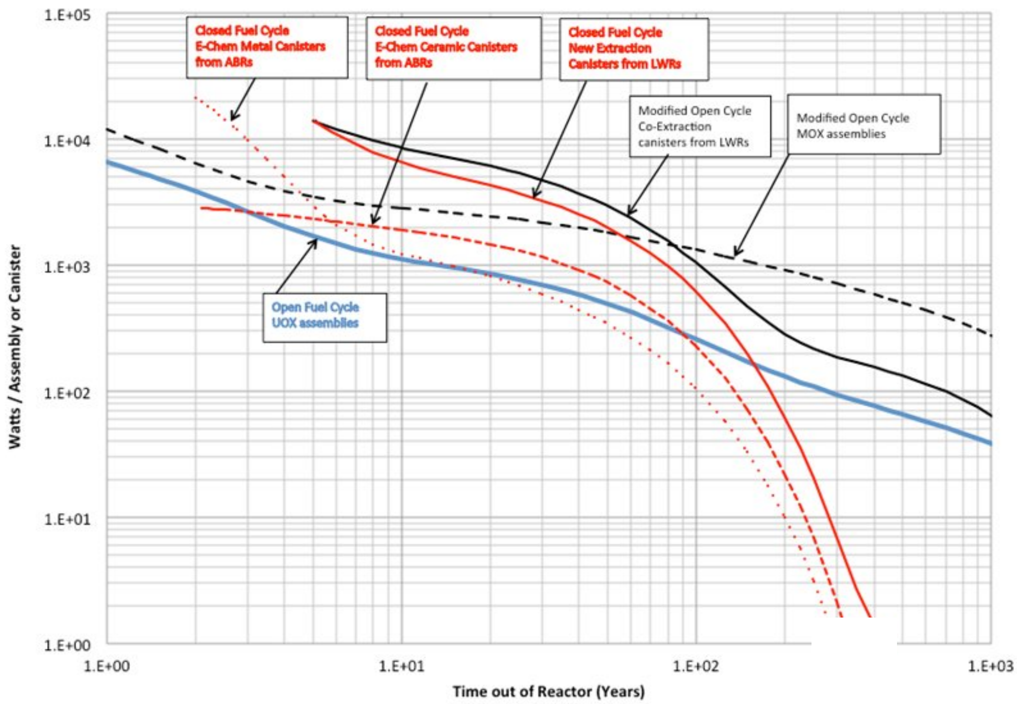

Figure 2 –Decay Heat Curves for 1 SNF Assembly or 1 HLW Canister

ANALYTICAL MODEL DEVELOPMENT

The transient "outside" model

The transient "outside" model was developed assuming a homogeneous infinite medium to portray the temperature transient in the host rock. A suite of analytical heat transfer model solutions for basic geometries was summarized in [3], based on methods and equations from Carslaw and Jaeger [7]. The following subset was chosen for the current analysis:

- A point source in an infinite medium, representing a single adjacent waste package (as a point source), where the point source heat load is the total heat source for a waste package
- A finite line source in an infinite medium, representing a central waste package of interest with a line load heat source internal to a single waste package
- An infinite line source in an infinite medium, representing an average line load of adjacent emplacement drifts or boreholes, where the line load heat source represents an average heat load accounting for axial waste package center-to-center spacing.

The unit cell for each model included 9 axial waste packages (4 on either end of the central finite line waste package heat source) and 9 adjacent drifts (4 on each side of the central drift.

The analytical solution equations applied are shown in Equations 1-3 below, and were derived in our earlier work [3]:

$$T_{point}(t,r) = \frac{1}{8\cdot\rho\cdot Cp\cdot(\pi\cdot\alpha)^{\frac{3}{2}}}\cdot\int_0^t \frac{q(t')}{(t-t')^{\frac{3}{2}}}\cdot e^{\frac{-r^2}{4\cdot\alpha\cdot(t-t')}}\,dt'$$

Equation (1)

The variable r in Equation (1) can be represented in the Cartesian coordinates of the repository, where the source is located at (x_0, y_0, z_0). For the finite line source, y_0 is integrated over the length of the finite line (with line length L), and the line source is centered at y=0. The definite integral over dy_0 is recognized as a form of the error function (erf) and the coordinate (x_0, z_0) is set to (0, 0).

$$T_{line}(t,x,y,z) = \frac{1}{8\cdot\pi\cdot k}\cdot\int_0^t \frac{q_L(t')}{t-t'}\cdot e^{\frac{-(x^2+z^2)}{4\cdot\alpha\cdot(t-t')}}\cdot\left[\operatorname{erf}\left[\frac{1}{2}\cdot\frac{\left(y+\frac{L}{2}\right)}{\sqrt{\alpha\cdot(t-t')}}\right]-\operatorname{erf}\left[\frac{1}{2}\cdot\frac{\left(y-\frac{L}{2}\right)}{\sqrt{\alpha\cdot(t-t')}}\right]\right]dt'$$

Equation (2)

$$T_{infinite\_line}(t,x,z) = \frac{1}{4\cdot\pi\cdot k}\cdot\int_0^t \frac{q_L(t')}{t-t'}\cdot e^{\frac{-(x^2+z^2)}{4\cdot\alpha\cdot(t-t')}}\,dt'$$

Equation (3)

This subset of solutions was then combined based on linear superposition in a conduction-only heat transfer environment (Duhamel's theorem, [7]).

To model the various repository designs in the different environments, the respective axial and lateral spacing used for each design are shown in Table I.

Table I - Repository Design Axial and Lateral Spacing (m)

Geology	SNF		HLW	
	Axial	Lateral	Axial	Lateral
Granite	10	20	10	20
Clay	10	30	6	30
Salt	20	20	20	20
Deep Borehole	6	200	6	200

<u>The quasi-steady-state "inside" model</u>

The temperature histories within the EBS (referred to here as the "inside calculation") are derived from the waste package line load heat source, using the time-dependent results of the “outside” calculation. The “inside” calculation is steady-state at each point in time, which is equivalent to assuming that the heat flow through the calculation radius at any given time is nearly equal to the heat generation in the waste at that time. This is a reasonable assumption except at the very early times in which the EBS temperatures are changing rapidly due to the change in boundary condition after emplacement. This calculation is conservative in the sense that the steady-state model is a one-dimensional model that effectively assumes an infinite line source with the waste package internal line loading.

The model for a multi-layer cylindrical steady-state temperature solution is derived from Kreith [8]. The derivation for use in our EBS-component models is described more in [3].

By conservation of energy at steady state, the temperature at the surface of each EBS component can be calculated a series of thermal resistance values based on the component radii and the thermal conductivities. Equation 4 calculates the temperature rise across the liner to give the surface temperature at the outer radius of the backfill layer.

$$T_{BACKFILL} = T_{DW} + \frac{q_L}{2\cdot\pi\cdot r_{DW}}\cdot R_{LINER} = T_{DW} + \frac{q_L\cdot r_{DW}\cdot \ln\left(\frac{r_{DW}}{r_{BACKFILL}}\right)}{2\cdot\pi\cdot r_{DW}\, k_{LINER}} = T_{DW} + \frac{q_L}{2\cdot\pi\cdot k_{LINER}}\cdot \ln\left(\frac{r_{DW}}{r_{BACKFILL}}\right)$$

Equation (4)

This equation and the approach were input into MathCad [9], and validated against example problems in Kreith [8].

The subscript DW derives from “drift wall”, but the variable is also used for borehole radius, or in some cases a “calculation radius” which may be some distance into the near-field rock, where a concentric layer of near-field rock is included as another internal layer. The “calculation radius” approach was applied in the salt environment to examine the effect of changing properties in a given layer of host rock. This was applied specifically for the salt repository concept, where there was a layer of crushed salt that consolidates over time and then has the properties of intact salt.

Modeling the Effects of Surface Storage Time

Surface storage times of 10, 50, 100, and 200 years were evaluated for all cases for the six different types of spent fuel and nuclear waste. The number of assemblies per package was varied from 1 to 12 based on known package designs. For deep borehole design concepts, due to the smaller diameter of the HLW canisters needed to fit in a borehole, the relative number of assemblies per waste package was 0.29.

The combination of heat sources and layout dimensions was used for the “outside” calculation to develop the transient temperature at the “calculation radius”, which corresponded to the host rock wall adjacent to the center of the finite line source. This transient rock wall temperature was then used in conjunction with the heat source of the waste package (represented by the finite line source) and the “inside” quasi-steady-state multi-layer model to develop the temperature gradient through the EBS components.

Additional Considerations

The current set of analytical models assumes constant thermal properties for the host rock, whereas some of the thermal properties, such as thermal conductivity and thermal diffusivity, are in reality functions of temperature, porosity, or moisture content that can vary over time. The properties of salt, in particular, demonstrate a strong dependence on temperature. This has not been addressed in the current analysis, except in an effort to bound the variation of salt properties by using crushed and intact salt properties.

Clayton and Gable [10] provide data addressing the thermal conductivity and diffusivity of intact salt with temperature and of crushed salt with porosity and temperature. They also provide a discussion of the time for reconsolidation of crushed salt to intact salt. Figure 3 was derived from the equations and data in [10].

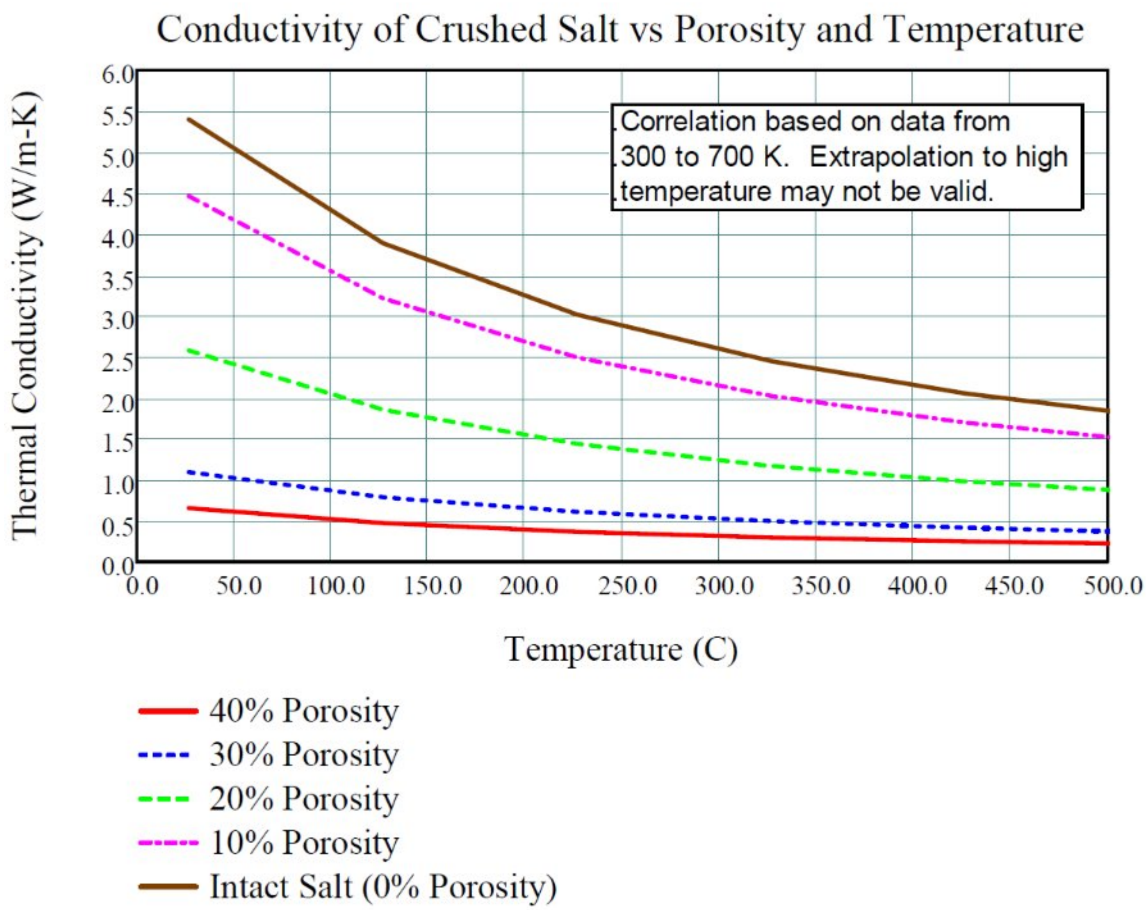

Figure 3 – Thermal Conductivity of Crushed Salt as a Function of Porosity and Temperature

RESULTS

In the transient "outside" calculation a constant intact salt thermal conductivity at 100°C was assumed for the host rock environment. To examine the effects of crushed and intact salt in the EBS on the waste package temperature over time, an example calculation was performed using a 4-assembly MOX waste package for crushed salt (35% porosity), intact salt, and a more realistic mixture of 25% crushed and 75% intact salt in the generic salt repository design [2], where the waste packages are placed horizontally in the bottom corner of an alcove excavated in salt, and then covered in crushed salt (Figure 4). Note that the results for crushed salt surrounding the waste package in the EBS is not realistic for several reasons – the model assumes a constant thermal conductivity based on 35% porosity and thermal conductivity at 100°C; consolidation effects are not accounted for; and salt melts at around 800°C [11], where thermal conductivity would increase significantly. Additional investigations with finite element codes are planned for FY12 to address this particular geometry, and to deal with variation of salt properties with temperature and consolidation of the crushed salt.

All of the results for the salt environment in the EBS presented in this paper are based on the case where 75% of the waste package surface was assumed to be in contact with intact salt.

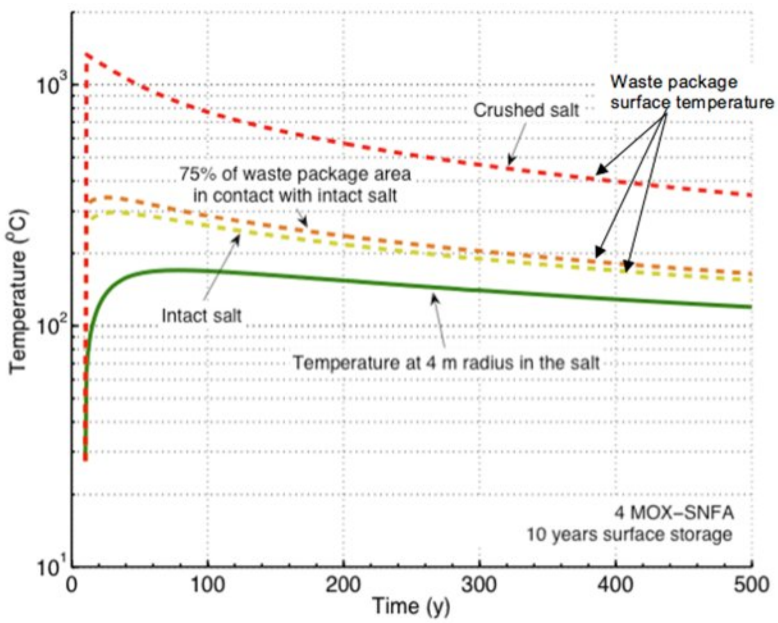

Figure 4 – Waste Package Surface Temperature for a 4-Assembly MOX Waste Package in Crushed Salt (35% porosity), Intact Salt, or a Combination (75% Contact with Intact Salt)

Figure 5 shows the transient temperature at the host rock interface for waste packages containing 1, 2, 3, 4, and 12 assemblies of UOX waste packages, for pre-emplacement surface storage times of 10, 50, and 100 years (shown as dot/dashed, dashed and solid lines respectively). Each figure includes four frames to present the results in granite, clay, salt, and deep borehole environments. The deep borehole environment has a higher ambient temperature than the other media due to the geothermal gradient (assumed to be 25°C per 1,000 m depth). The figures show unsurprisingly that minimizing the waste package capacity and maximizing surface storage time results in lower rock wall temperatures. It is important when designing the repository to not exceed the thermal constraints of the EBS components or the geologic media.

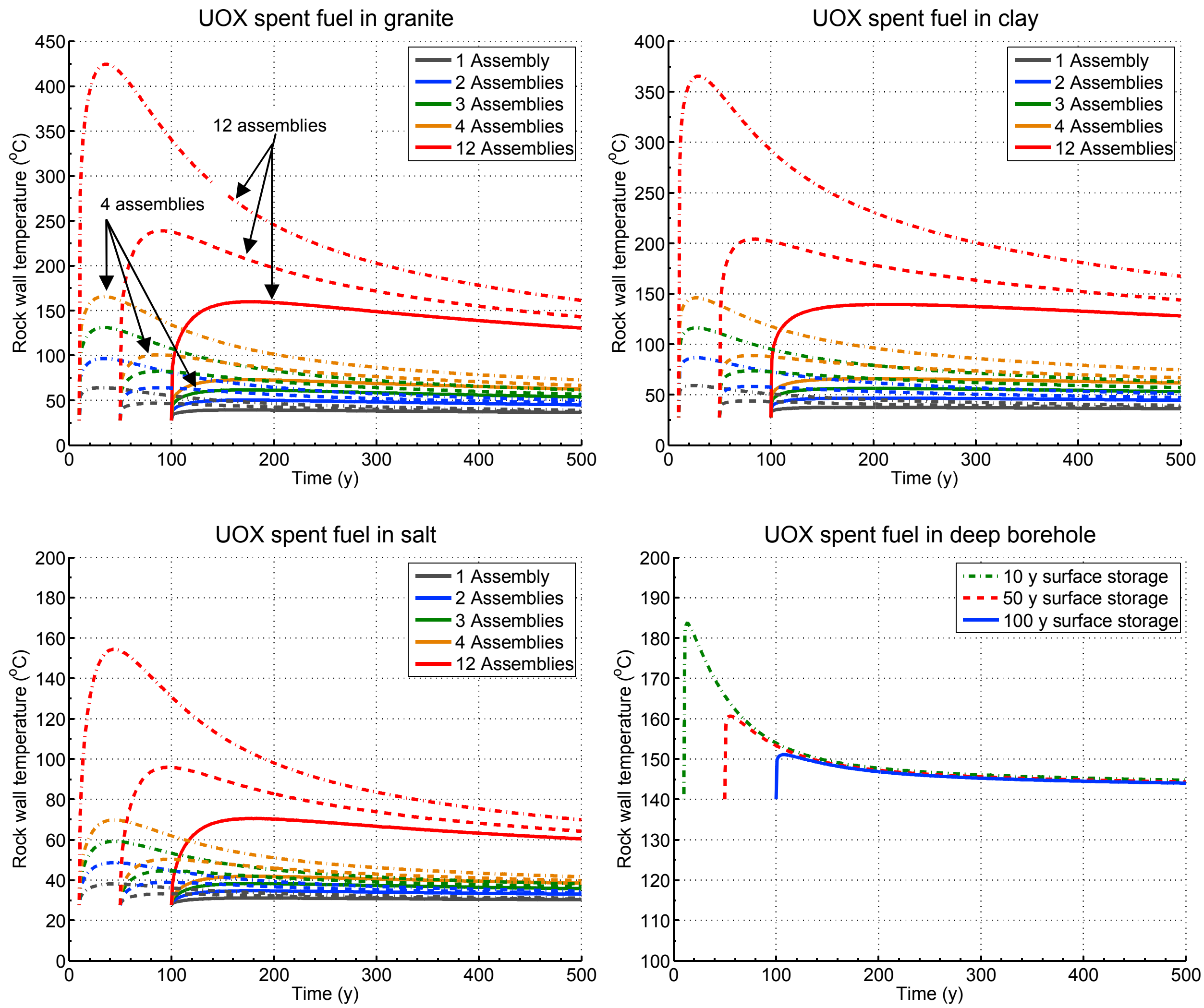

Figure 5 – Temperature Transients for 1, 2, 3, 4, and 12 Assembly Waste Packages of UOX in Granite, Clay, and Salt; and 1 assembly Rod Consolidation WP in a Deep Borehole

Figure 6 shows equivalent transient temperatures of MOX waste packages in each of the 4 different geologic media and different storage times. The results show significantly higher temperatures are obtained compared to UOX waste packages, which is expected given that a MOX waste package typically contains approximately 3 times more heat than its UOX equivalent. The results also show that some combinations of MOX waste packages and geologic media may not be feasible with respect to the thermal constraints.

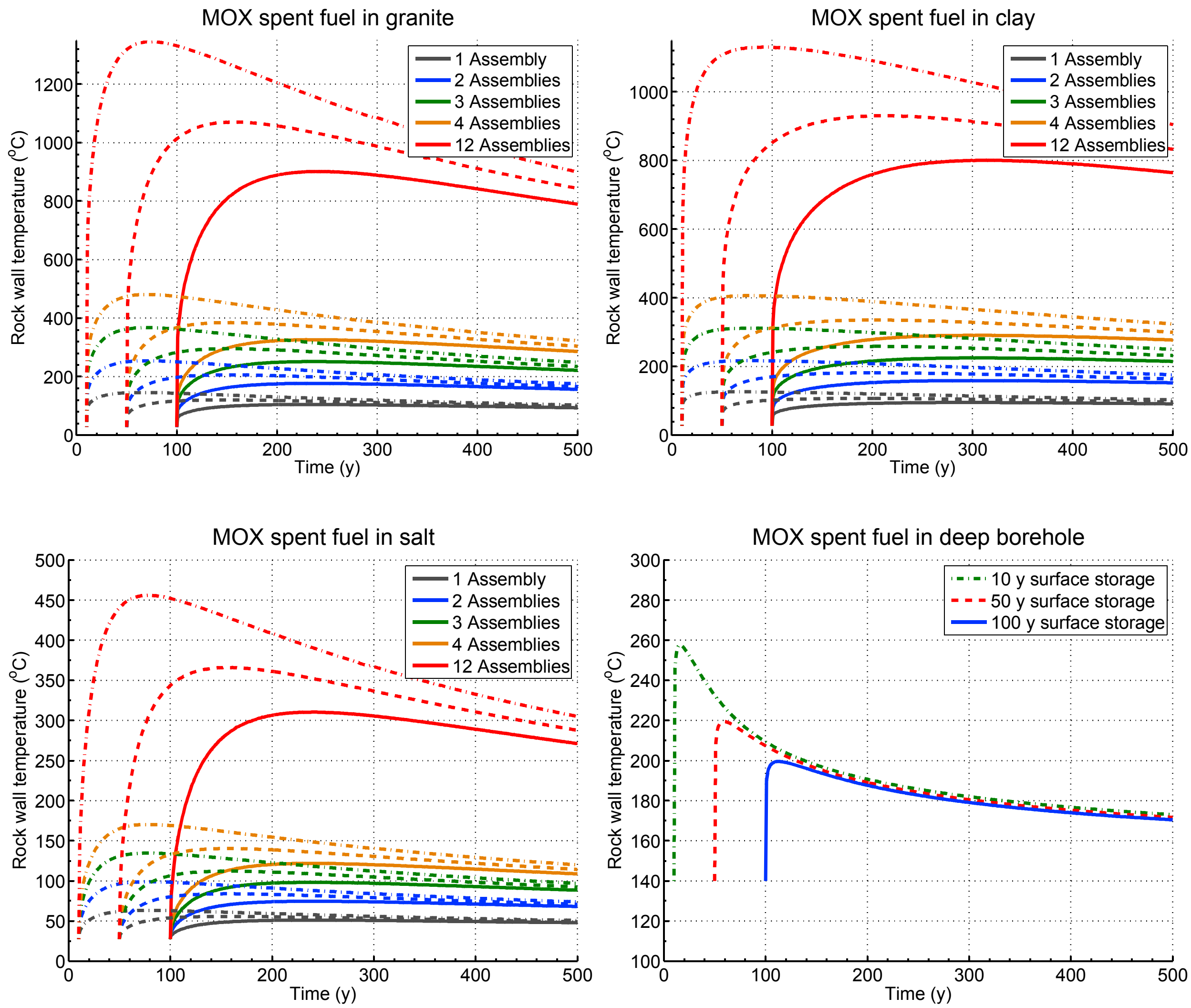

Figure 6 – Temperature Transients for 1, 2, 3, 4, and 12 Assembly Waste Packages of MOX in Granite, Clay, and Salt; and 1 Assembly Rod Consolidation WP in a Deep Borehole

Figure 7 shows the transient temperature at the host rock interface for all four types of HLW canisters (Co-extraction glass, New-extraction glass, E-chem ceramic, and E-chem metal) in granite for 10, 50, and 100 years of surface storage. Similar models were calculated for clay, salt and deep borehole (although not shown here). It is again apparent that maximizing pre-emplacement surface storage prevents temperatures from exceeding the thermal constraints at the rock wall. HLW glasses generate more heat than the electrochemical ceramic and metal wastes.

As described earlier, the overall repository unit cell thermal behavior is a combination of a central waste package finite line source, neighboring axial waste package point sources, and adjacent drift/borehole infinite line sources.

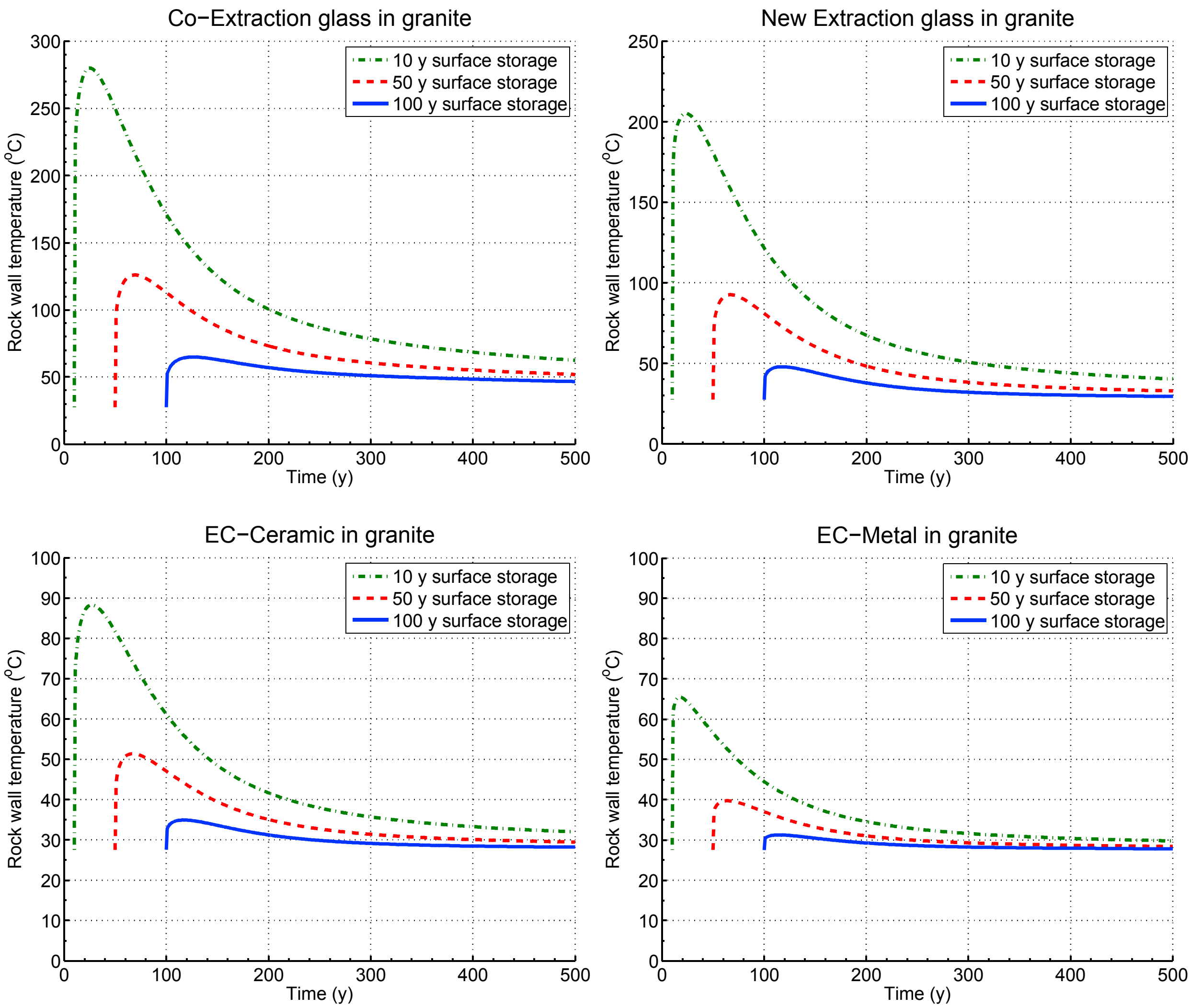

Figure 7 – Temperature Transients for HLW Canisters of Co-Extraction Glass, New-Extraction Glass, E-Chem Ceramic, and E-Chem Metal in Granite

Figure 8 shows the temperature transient contributions to the rock wall temperature from the central waste package, the adjacent waste packages in the same line, and the adjacent drifts for a 1-assembly UOX waste package in granite, clay, salt, and deep borehole environments. This figure shows the dependence of the different contributions on waste package and drift (or borehole) spacing. In granite, the central waste package dominates the temperature at the rock wall until approximately 40 years after emplacement, when the adjacent drifts dominate. Similar is true for clay and salt, where the central waste package dominate for the first 75 and 20 years respectively. For deep borehole, the central waste package dominates beyond 500 years.

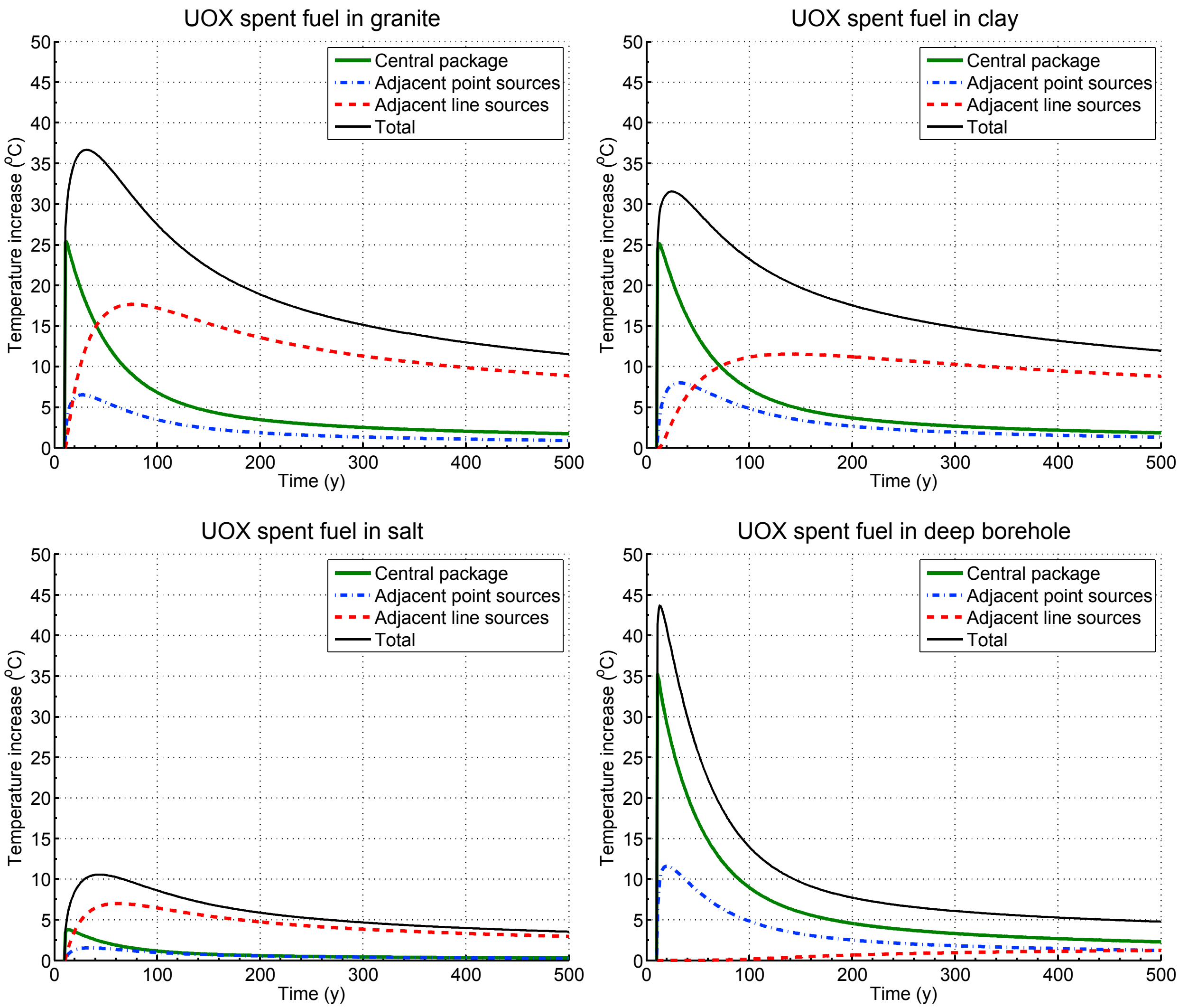

Figure 8 – Temperature Transient Contributions from the Central Waste Package, the Adjacent Waste Packages in the same line, and the Adjacent Drifts for a 1-Assembly UOX Waste Package in Granite, Clay, Salt, and Deep Boreholes

The temperature at the waste package surface is undoubtedly higher than that at the rock wall, and thermal constraints may be more difficult to achieve closer to the waste package surface. Figure 9 shows waste package surface temperature transients for a 1-assembly UOX waste package in granite, clay, salt, and deep borehole environments. The temperature at the waste package can be reduced by increasing the surface storage time, or by increasing the thermal conductivity of some of the EBS material layers between the waste package and the drift wall.

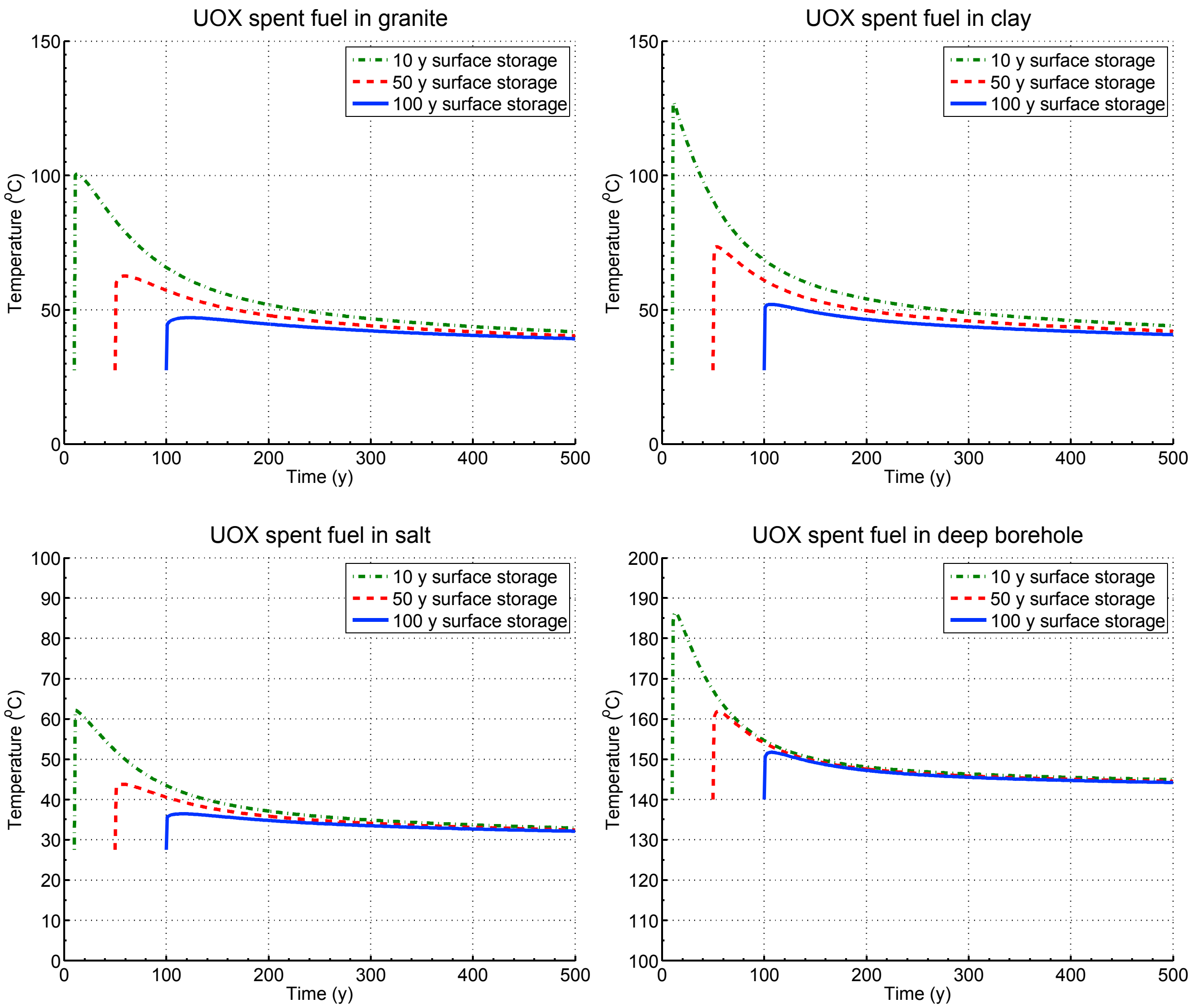

Figure 9 – Waste Package Surface Temperature Transients for a 1-Assembly UOX Waste Package in Granite, Clay, Salt, and Deep Borehole

To more closely examine the effect of surface storage time on the peak waste package temperature of both UOX and MOX waste packages of different capacities, Figure 10 shows peak waste package surface temperature transients for 1, 2, 3, 4, and 12 assembly waste packages of UOX and MOX in granite and clay as a function of surface storage time. The results show that 4-aseembly UOX waste packages can be placed in a granite or clay repository after just 100 years of surface storage without exceeding the thermal constraint for clay or bentonite buffer. For MOX however, significant storage time is required for any packages larger than one assembly in granite and clay.

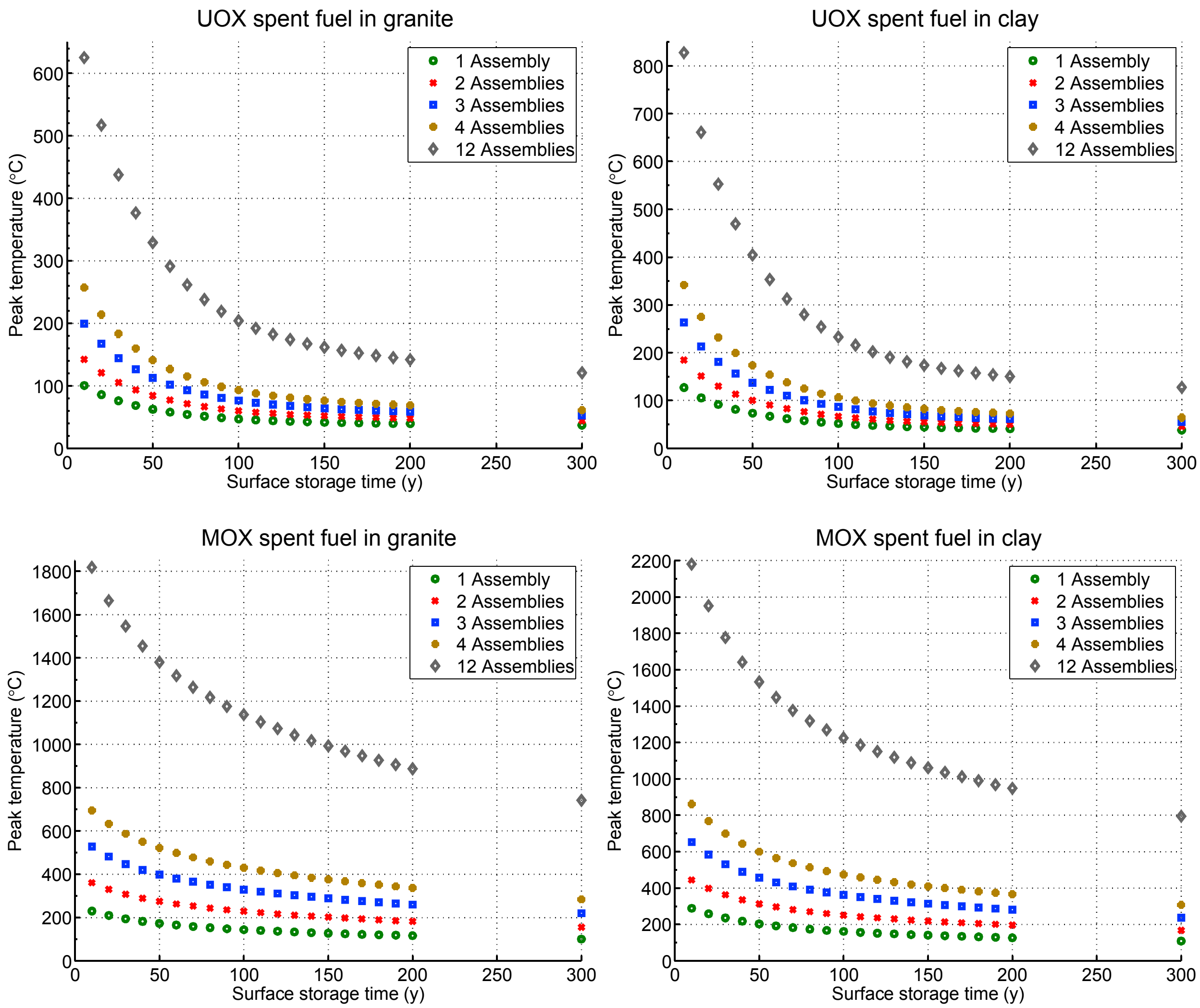

Figure 10 – Peak Waste Package Surface Temperature Transients for 1, 2, 3, 4, and 12 Assembly Waste Packages of UOX and MOX in Granite and Clay

The storage time required to meet the thermal limits of the EBS components for UOX and MOX in granite, clay and salt are shown in Figure 11. The constraints are 100°C for clay or granite (with bentonite buffers), and 200°C in salt. The 100°C temperature constraint was chosen to limit alteration of clay in buffers, for example by illitization or cementation. Alteration generally involves dissolution, aqueous transport, and precipitation. For salt, a more ductile material, a higher target value of 200°C is used for the maximum temperature, to limit uncertainty in performance assessment. Additional thermal constraints for different materials such as metal liners may exist.

Based on these results, up to 4 assemblies of UOX SNF can be emplaced in any of the media with approximately 100 years or less of surface storage. A 12-assembly UOX waste package requires around 50 years of surface storage for emplacement in salt, and about 500 years or more for emplacement in clay or granite. Similarly for MOX (which, as shown in Figure 2, is initially around 3 times as hot, but stays hot much longer than

UOX assemblies), 4 assemblies can be disposed of in a salt repository after around 100 years of surface storage. In granite and clay repository concepts a single assembly of MOX would require around 300 years of surface storage, for the repository layout parameters evaluated, to meet the thermal constraints assumed.

Since the storage time is driven largely by the material properties and thermal constraints of the engineered and natural barriers, the effects of relaxing the thermal constraints were investigated. It is again important to note that these thermal constraints are preliminary, and subject to change based on site-specific data and further studies. Variations on clay buffer limits have been proposed, for example, limiting an outer portion of the buffer cross section to 125°C [12]. Furthermore, for a salt repository, a thermal constraint of 250°C was adopted for the Deaf Smith County design concept [13].

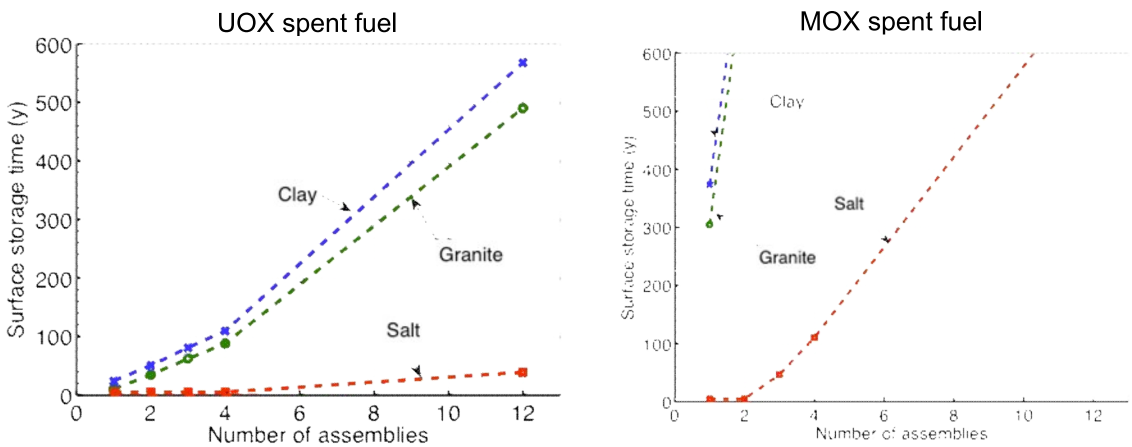

Figure 11 – Surface Storage Time Required to Comply with Temperature Constraints of 100°C for Clay or Granite (with Bentonite Buffer), and 200°C in Salt

Figure 12 shows the sensitivity of required surface aging times of UOX and MOX waste packages to variations in the thermal constraints for granite and clay (100°C, 125°C and 150°C), and salt (200°C, 225°C, and 250°C). The results show that for UOX in granite and clay, shorter surface storage times are needed by relaxing the thermal constraints by just 25 – 50°C. Alternatively, higher capacity waste packages could be disposed of in some cases without significantly increasing the surface storage time. Because of the relatively high thermal conductivity of salt coupled with the much higher initial temperature constraint, the impact in salt is not as great as that observed in clay and granite. For MOX waste packages in granite and clay, the effect is negligible simply because no more than one assembly can be disposed of after even 300 years of surface storage even with the relaxed thermal constraints. For MOX in salt, relaxing the thermal constraints by 25 or 50°C may allow larger capacity waste packages or shorter pre-emplacement storage times to be used.

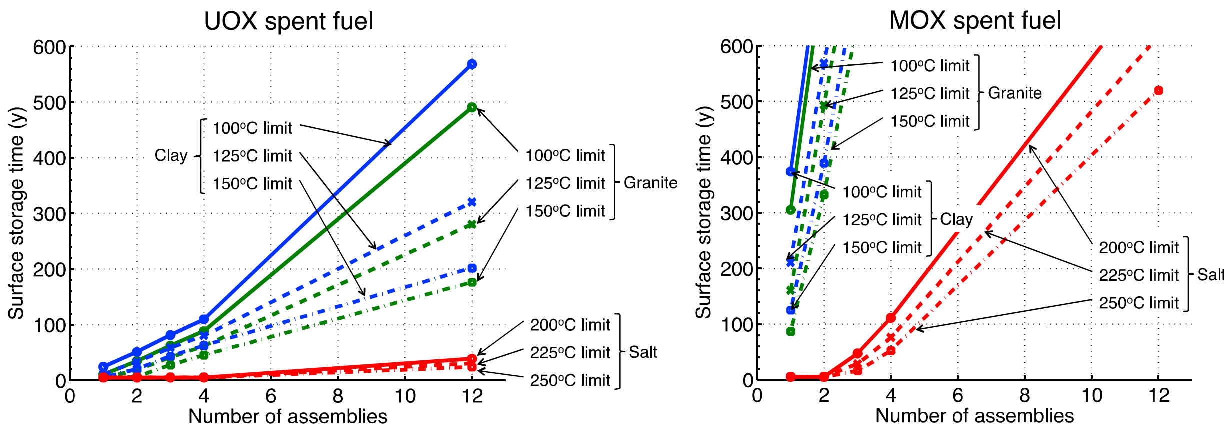

Figure 12 – Surface Storage Time Required for Compliance with Temperature Constraints of 100°C, 125°C, or 150°C for Clay or Granite (with Bentonite buffers) and 200°C, 225°C, or 250°C for Salt

Figure 13 shows the effect of changing the thermal conductivity of the buffer layer of the EBS from 0.6 W/m-K for a dry bentonite buffer, to 2.0 W/m-K on required surface storage time for UOX and MOX in granite and clay repositories. Increased thermal conductivity of the buffer layer can be achieved by using an engineered buffer consisting of a mixture of graphite, sand, and bentonite. Examples of such mixtures are presented in [14].

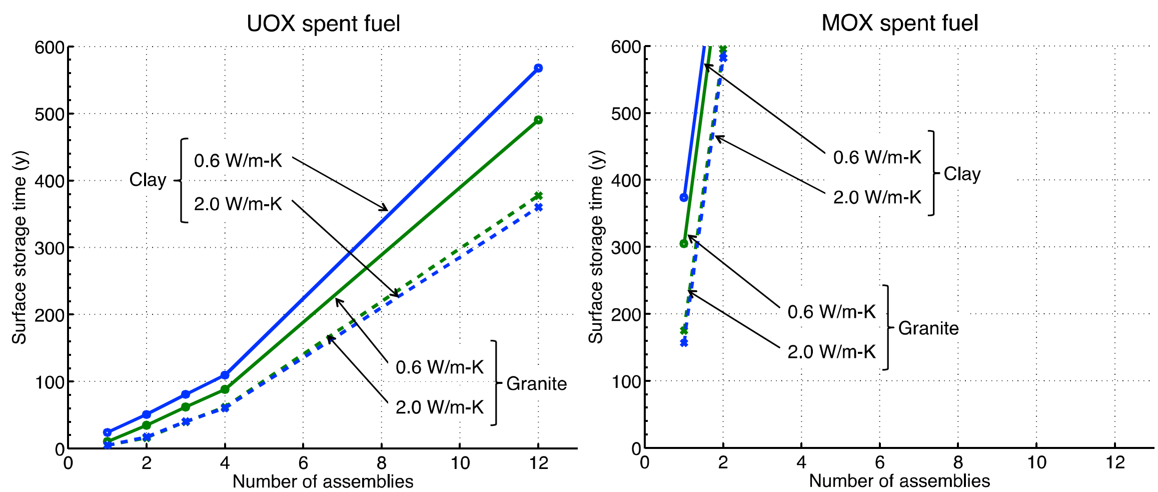

Figure 13 - Effect of Changing the Thermal Conductivity of the Bentonite Buffer Layer in the Repository Designs for Granite and Clay

CONCLUSIONS

The analytical solution approach used to analyze the repository temperature response allowed rapid comparison of a large number of alternative disposal options and design configurations. More than 100 combinations of waste form, geologic environment, repository design configuration, and surface storage times were analyzed and compared. This approach allowed investigation of the sensitivity of the results to combinations of parameters that show that there is much flexibility to be gained in terms of spent fuel management options by varying a few key parameters.

This initial analysis used representative design concepts and thermal constraints based on international design concepts, and it also included waste forms representing future fuel cycles with high burnup fuels. Unlike repository designs with large open tunnels and pre-closure ventilation, all of the disposal concepts analyzed in this study used enclosed emplacement modes, where the waste packages were in direct contact with encapsulating engineered or natural materials.

The deep borehole repository concept limits the size of the SNF waste packages and may require rod consolidation to fit within the drill casing diameter. A single assembly waste package, assuming rod consolidation, was evaluated in the current analysis. Similar size restrictions apply for the HLW canisters. At this time no thermal constraints have been defined for the deep borehole repository concept.

Representative EBS materials and properties were evaluated. However, changes in EBS design concepts and materials can also have significant effects on the maximum waste package surface temperature. Increased thermal conductivity of the buffer layer can be achieved by using an engineered buffer consisting of a mixture of graphite, sand, and bentonite [14].

One of the advantages of the analytical model is that it highlights the sensitivity of the results to the parameters that define the repository layout, including spacing between axial and lateral neighboring waste packages and drifts. It is clear that repository layout adjustments can be made to reduce the calculated peak temperatures. The results also show that significant reductions in required surface storage times can be achieved if higher thermal constraints can be justified

Additional studies are planned to evaluate the trade-offs between surface storage times, repository layout parameters, and variations in EBS design concepts. Model validation and uncertainties will also be addressed. It is expected that shorter surface storage times and more optimized repository design configurations may be achieved.

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Disclaimer

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NOMENCLATURE AND SYMBOLS

Cp	specific heat, J/Kg-K
k	thermal conductivity, W/m-K
L	characteristic length, m
q(t)	continuous heat generation rate of the point source, W
$q_L(t)$	continuous line heat source, W/m
r	radial distance, m
R	thermal resistance, m-K/W
t	time, s
t'	integration variable for convolution integral
T	temperature, K
α	thermal diffusivity, $m^2/s = k/(\rho\text{-}Cp)$
ρ	density, Kg/m^3

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