Cleaning up Hanford: A Deep Borehole Disposal Concept for the Cs/Sr Capsules - 16249

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ABSTRACT

The Hanford site in Washington State is possibly the most radioactive-contaminated site in the USA presenting a major environmental problem requiring urgent clean-up. Sealed capsules containing halides of strontium (Sr) and cesium (Cs) account for over a third of the total radioactivity. Interim storage for a period of 10-half-lives is an unappealing option since many of the capsules contain Cs-135, which has a half-life of 2.3 million years, and the dose received from the shorter-lived isotopes would remain dangerously high for several generations. Deep geological disposal of the capsules would be a safer solution.

We discuss a new and revised conceptual model for disposal of the entire Hanford capsule inventory in a *single* deep geological borehole with a diameter that could be as little as 31 cm. The concept involves placing thirty capsules inside a steel-disposal container. Holes up to this size are regularly drilled to depths much greater than 5 km by the hydrocarbon industry and are well within current drilling capabilities. Numerical modeling work demonstrates the feasibility of the conceptual model. In particular we examine the thermal envelope generated as a function of time enabling decisions to be made on possible fillings for the borehole annulus. Variations on the disposal concept include disposing different combinations of capsules to control the heat output and thus the temperature rise. The results lend support to a comment attributed to Secretary Moniz that the Hanford capsules could be the world's first HLW to be disposed of in a deep borehole.

INTRODUCTION

As a legacy of the cold war, the Hanford site in Benton County, Washington is now the most radioactively contaminated site in the USA. Prefabricated capsules containing solid strontium fluoride (SrF_2) and cesium chloride CsCl) waste account for over a third of the total radioactivity at the site. The Hanford capsule inventory comprises 1935 capsules each with a length between 0.51 and 0.53 m (~20-21 in) and outside diameters of around 0.067 m (2.7 in). These capsules have a high initial thermal loading, though much of this will be exhausted over a time period of 200 years. As of August 29, 2007, the cesium capsules had a range of initial heat outputs of 93.86 – 195.37 W per capsule while the strontium capsules ranged from 22.12 – 504.63 W [1]. In terms of activity, the capsules contain the fissile isotopes: Cs-137 and Sr-90, having half-lives of 30.02 years and 28.79 years respectively. However, most of the cesium capsules contain in addition, a small amount of Cs-135, which has a half-life of 2.3 million years.

The existence of the capsules presents a serious environmental and societal problem requiring a solution. Two possibilities that have been mooted are: interim storage and geological disposal. The former entails placing the capsules in secure dry storage for a period of time of at least 10 half-lives by which time the heat output and radioactivity will be greatly reduced but this still leaves the question of a final disposal unanswered. Furthermore, the presence of the Cs-135 in the Cs capsules undermines the case for that subset of the Hanford inventory. The US DOE is currently considering disposing of the entire Hanford capsule inventory in deep geological boreholes. Indeed, US Secretary of Energy Moniz is reported to have said that the Hanford capsules could be the world's first HLW to be disposed of in a deep borehole. The deep borehole disposal (DBD) solution is appealing because the small size of the capsules means that the entire inventory could be disposed of in a single 5 km deep borehole using less than a 1 km disposal zone. Furthermore, the diameter of such a borehole is well within current drilling capabilities for vertical holes drilled to that depth and narrower than the Deep Borehole Field Test (DBFT) and pilot boreholes due to be drilled in late 2016 [2,3]. DBD offers superior safety over mined, engineered repository systems such as those being considered and pursued by several countries including France, Sweden, Finland, Switzerland and the UK for spent fuel and HLW. The extra safety is provided by an order of magnitude increase in the geological barrier and a stable system of saline groundwater [4]. An additional attraction of this method of disposal is the relative speed of implementation. A 5 km borehole at the required diameter could be drilled, cased and filled within a period of 2 years.

In this paper we build on our earlier research in which we introduced a baseline concept for DBD of the Hanford capsules using a container holding two capsules arranged axially, end to end, and requiring a 0.216 m (8.5 inch) diameter borehole. We introduce here a new "triples" concept in which we consider a disposal container housing 30 capsules, arranged in 5 rows of 3 end to end pairs of capsules requiring a 0.311 m (12.25 inch) diameter hole. Using numerical modeling, we demonstrate that the thermal envelope around a disposal zone using a single container of cesium-containing capsules allows the use of a superior sealing and support material – the High Density Support Matrix (HDSM) [5]. The use of this filling material removes one of the main potential weaknesses leveled at DBD: the possibility of radionuclide transport back to the surface via groundwater present in the annuli between the waste packages and the borehole wall. It also provides mechanical support and retards the ingress of groundwater which can cause container corrosion and eventual radionuclide escape [6].

METHODOLOGY

Baseline Disposal Concept

The Hanford capsules consist of a container in which CsCl or SrF_2 is sealed within inner and outer steel walls. The capsules vary in length between 0.51 and 0.53 m, while their diameters range from 0.067 to 0.083 mm. In two previous publications [1,7] we considered a baseline disposal case in which two capsules, axially aligned, are placed base-to-base in a 1.083 m long stainless steel container (henceforth referred to as the disposal container) with an outside diameter (O.D.) of 0.114 m and a wall thickness of 12.7 mm. This required a 0.216 m (8.5 in) diameter borehole and 0.178 m (7 in) O.D. casing (Figs. 1 and 2).

The void space between the capsules and container is filled with silicon carbide, a material with high thermal conductivity. The use of such an insert is necessary to minimize any risk of deformation or collapse under the disposal pressure and to ensure efficient conduction of decay heat away from the capsules. Silicon carbide is inexpensive, lightweight and can easily be machined with special “cut-outs” to accommodate the capsules within the container. The annuli between the container and casing and between the casing and host rock should ideally be filled with a sealing and support matrix (SSM). The primary role of an SSM is to inhibit the access of groundwater to the casing and disposal containers for as long as possible, thereby preventing or delaying any corrosion and closing a possible escape route for any gaseous corrosion products or released radionuclides back to the surface.

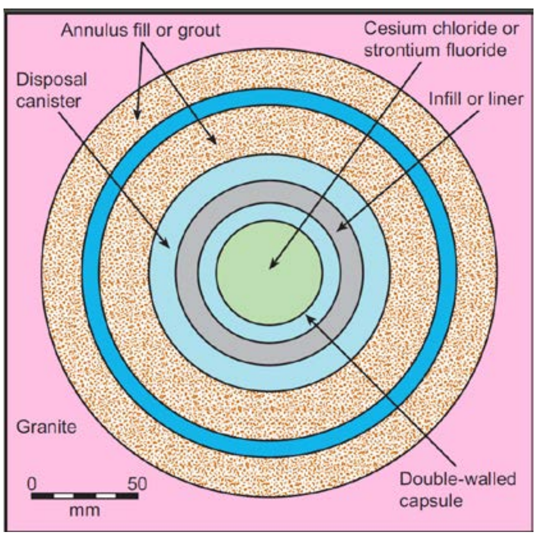

Fig. 1. Horizontal cross section of “baseline” DBD concept for CsCl capsules. The outer (darker blue) ring is the casing.

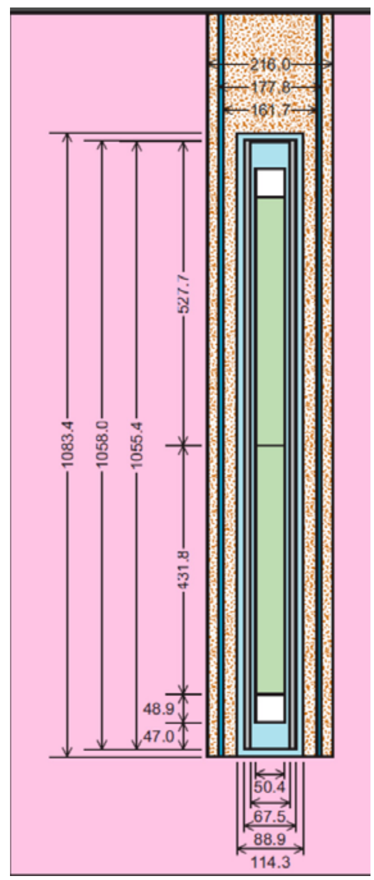

Fig. 2. Vertical cross sections of the “baseline” DBD concept as simplified for thermal modeling.

Their secondary function is to provide mechanical support against the hydrostatic pressure in the disposal zone and axial load stresses from the overlying waste containers, especially in the period before the borehole is sealed. In the longer term an SSM could also help to counteract any tectonic stress on the casing and waste packages.

In our previous work, where we considered only the 2-capsules-per-container baseline case, an SSM was not really necessary given the small size and low weights involved. However, it is still important to fill the annuli and we considered bentonite for this purpose (without specifying how this filling material would be emplaced). The baseline DBD concept would require a stack of containers having a total height of ~ 1 km (2 capsules per 1m container for the total inventory of 1935 capsules). In References [1, 7] we suggested that more efficiency might be attained if wider boreholes were considered, allowing a higher packing of capsules in the disposal containers. For a hole 0.311 m (12.25 in) in diameter, we proposed placing 6 capsules per container in two rows of three. In the present work, we extend this idea, but increase the number of capsules by considering a taller disposal container.

“Triples” Concept

Based on a borehole 0.311 m (12.25 in) in diameter within the disposal zone, and borehole casing of O.D. 0.272 m (10.7 in), we propose a stainless steel disposal container with an O.D. of 0.195 m, wall thickness of 16 mm and a (outside) height of 5.317 m. With these dimensions, we can fit 5 rows of 3 double capsules – 30 in total (see Figures 3 and 4 for vertical and horizontal cross sections including all measurements).

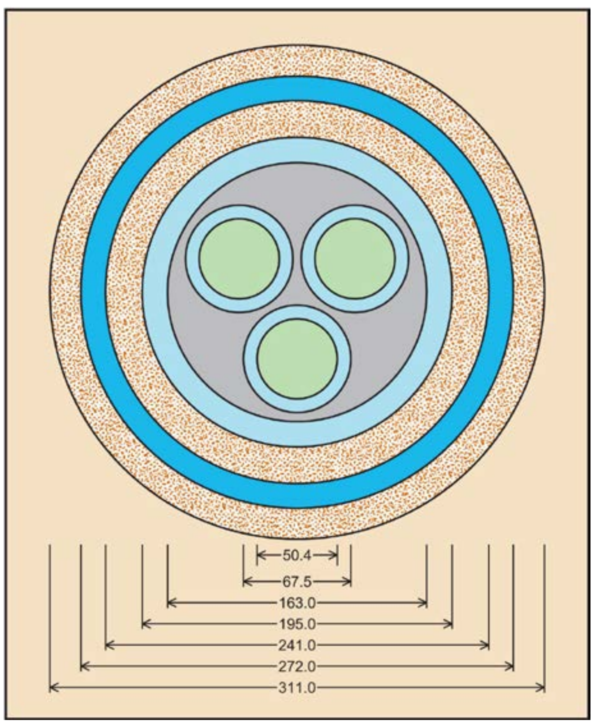

Fig. 3. Horizontal cross section of “triples” DBD concept for CsCl capsules. Key as in Fig. 4.

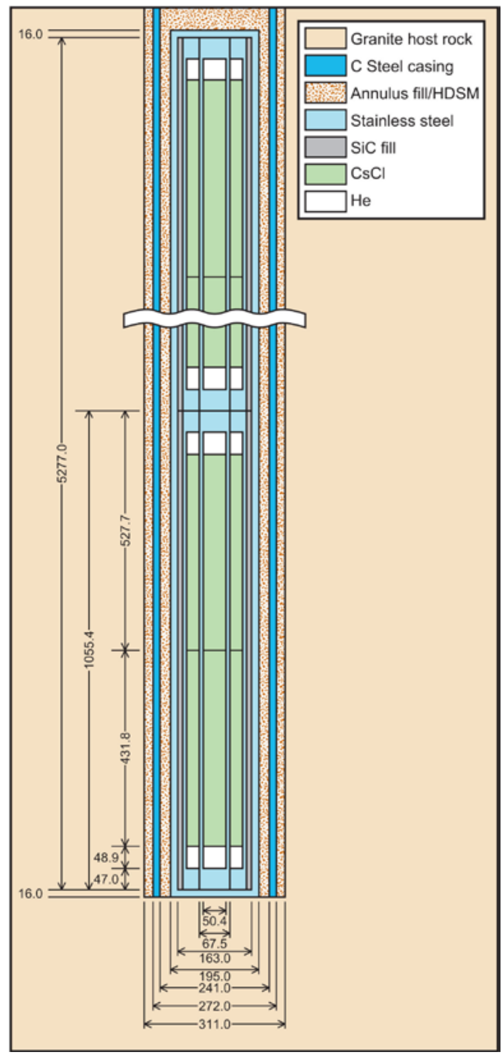

Fig. 4. Vertical cross section of the “triples” DBD concept as simplified for thermal modeling. The cross section is viewed from a point just inside the front of the container.

Because the triples DBD concept involves much larger containers and a much heavier package weight than for the baseline case, we propose using HDSM as an SSM [5,6]. HDSM is a fine, lead-tin (Pb-Sn) alloy shot which can be emplaced easily via drill string or coiled tubing.

Shot particles flow easily down the sides of the containers to fill the inner annulus and, if perforated casing is used, also into the outer annulus. Radioactive decay heat from the waste would result in melting of the alloy to form a dense liquid that will fill all remaining spaces, including some of the larger fractures in the wall rock. This can take as little as a few days in the hottest region of the annulus, near the center of the container. A typical Pb-Sn alloy with a composition close to the binary eutectic will melt around 185°C at one atmosphere (and only a few °C higher at disposal zone pressures). Once the decay heat subsides, this molten metal will cool and re-solidify, effectively soldering the waste into the borehole, and thereby providing a superior sealing function while at the same time offering mechanical support. Re-solidification occurs over a time span from a few years to a few decades depending on the maximum temperatures attained.

The triples DBD concept for the Hanford capsules is appealing for several reasons. First, the entire inventory of capsules could be disposed of with a disposal zone less than 400 m in length – a 60% reduction over the baseline concept. Second, the width of borehole, while 50% greater than that used for the baseline concept, is still narrower than the diameter of the DBD Field Test borehole. Third, any increased costs incurred by the need to drill a wider hole are likely to be offset by the smaller number of disposal containers required to be fabricated and the shallower disposal zone which would mean the depth of the borehole could be shortened to 4 km instead of 5 km. In this paper we concentrate on modeling the disposal of caesium containing capsules only, for it is these capsules that represent the greatest health risk due to the presence of the Cs-135 isotope, while the strontium capsules would merely add to the heat output.

Numerical Heat flow Modeling

The 3D thermal envelope surrounding one or more disposal containers may be obtained by solving the following heat conduction equation (Eq. 1):

$$\frac{\partial^2 \boldsymbol{T}}{\partial \boldsymbol{t}^2} = \boldsymbol{\alpha}\, \nabla \cdot \left(\boldsymbol{\kappa} \frac{\partial \boldsymbol{T}}{\partial \mathbf{r}} \right) + \sigma \quad \text{(Eq. 1)}$$

where T is the temperature, $\boldsymbol{r}$ is the spatial position, α is the thermal diffusivity, κ is the thermal conductivity and σ is the source term. We have assumed, following Hodgkinson [8], that conductive and convective heat flow can be decoupled and therefore treated separately. In this work we consider only the conductive flow of heat. For the particular case of DBD under consideration, the cylindrical symmetry can be exploited to simplify Eq. 1, yielding (Eq. 2):

$$\frac{1}{R}\frac{\partial}{\partial R}\left(\kappa R\frac{\partial T}{\partial R}\right)+\frac{\partial}{\partial z}\left(\kappa\frac{\partial T}{\partial z}\right)=\frac{\partial T}{\partial t}-\frac{S}{\rho c} \quad \text{(Eq. 2)}$$

where *R*, *Z* are the respective radial and vertical coordinates, *ρ* the density of the source term, *c*, its specific heat and *S* is the volumetric rate of heat production. Eq. 2 may be solved using the method of finite differences [9]. In our work we have used a non-uniform mesh; the mesh spacing is finer nearer the source. Mesh points are placed in five different materials: the contents of the disposal container, the disposal container material (stainless steel), the borehole filling material (HDSM), borehole casing (mild steel), and “granite” rock. For each of these different materials, temperature dependent thermal conductivities, specific heats and densities have been used. The internal contents of the container require further explanation. The container will hold one or more capsules, themselves consisting of CsCl, stainless steel capsule material, helium and steel end-caps (see Figs. 1-4). The remaining space in the container is filled with silicon carbide. Thermophysical properties of this “composite” material (everything inside the disposal container) are obtained using well-known mixing rules (density and specific heat) or a Kelvin-Voigt model for combining component thermal conductivities [7].

For the source term, decay heat data for Cs have been obtained from [1]. Cubic splines have been used to interpolate between successive tabulated data points. We considered a disposal in the year 2025. Initial heating rates at the year 2025 (note that the baseline case [1, 7] used 2020 outputs) vary between 63.018 W and 131.173 W per capsule. These values were converted to initial volumetric heating rates by first multiplying by 30 (for the “triples” arrangement), and then dividing by the internal volume of the disposal container, yielding average content values of 17.816 Wm^{-3} and 37.085 Wm^{-3} respectively.

The finite difference runs were conducted using a constant timestep of 500 seconds and covering a maximum timespan of 5000 days, depending on the runs conducted. The disposal container was considered to be in place at the bottom of the 5 km borehole but at “well-head” temperature at the start of the calculations. No allowance was made for any change in temperature during emplacement. Whilst in a real disposal situation, the temperature would increase during emplacement, any such changes might only affect the outer parts of the package if coiled tubing was used to ensure a rapid descent of around 2-3 hours. The set of points: {0,R=0.001m} represent the left hand edge of the mesh, which we henceforth refer to as the borehole axis. The 1 mm offset avoids mathematical difficulties when *R* = 0. The borehole bottom “ambient” temperature is taken to be 100°C. For temperature-time plots, temperatures are reported for radial positions at 3 fixed vertical distances: {0,R}, {H/2,R} and {H,R} that is, at the bottom of the disposal container, at its midpoint and at the top (when more than one container is disposed, the center of the stack is used for the midpoint). The chosen radial positions are on the borehole axis, at points within (i) the “waste”, (ii) the container wall (iii) the annulus between the container and the casing, (iv) the casing, (v) the annulus between the casing and the borehole wall, and at various points within the host rock.

RESULTS AND DISCUSSION

For our “triples” concept, based upon filling a container with 30 capsules of CsCl, all with the minimum initial heat output, the variation of temperature with time can be seen in Fig. 5 (Fig. 6 is the plot for the baseline concept - a single container with a pair of caesium capsules - added for comparison purposes). The results for this case show that the temperature rises to a maximum at around 580 days at the center of the container ie. less than 2 years post disposal. The hottest temperature achieved is ~203°C – on the borehole axis at a height midway up the disposal container. Very little temperature difference is observed between a point at the container outer surface and at the borehole wall. This observation reflects the use of the HDSM, which being metallic, has a high thermal conductivity, facilitating the transport of heat away from the source.

For this low heat output case, sufficient heat is supplied from the waste to reach the solidus of the HDSM around the center of the disposal container. However, this is not the case at the top and bottom of the container, where the HDSM would remain in a solid state. By using a mix of capsules, in which capsules with a higher initial heat output are added in place of this hypothetical case of 100% with the lowest initial output, a more complete melting of the HDSM in the annulus could be achieved.

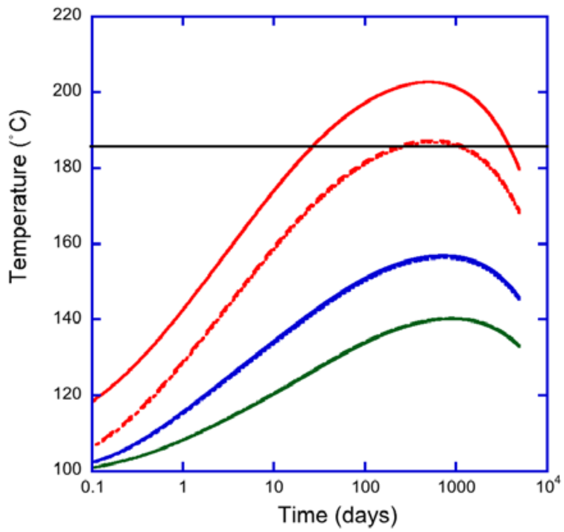

Fig. 5. Temperature versus time plot for “triples” concept using minimum heat output for Cs (see text). Key: red line: midway up container;blue: bottom of container;green: top of container. Solid lines: borehole axis; dashed lines: container surface; dotted lines: borehole wall. Horizontal black line : 1 atm HDSM solidus HDSM (185°C).

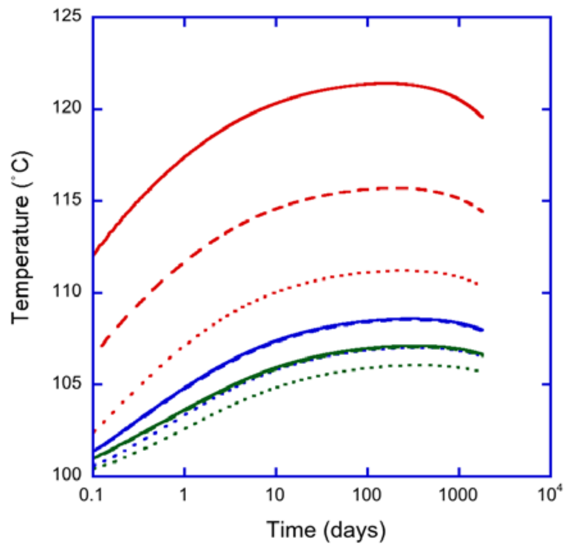

Fig. 6. Temperature versus time plot for baseline concept using minimum heat output for Cs (see text). Key – as for Fig. 5.

The thermal "footprint" of a triples disposal case can be ascertained by plotting the peak temperature [10] calculated over the course of a simulation at various radial positions taken at three different vertical heights corresponding to the top, bottom and centre of the disposal container. Fig.7 gives the results taken from a disposal container filled with capsules all with the minimum initial heat output (and for comparison purposes, Fig. 8 shows a similar one-container plot but for the baseline case). As the results show, the thermal footprint is very small; temperature rises to less than 4°C above ambient within a distance of only 10 m from the borehole axis, although this is still greater than for the baseline case.

The result of replacing all the CsCl capsules with minimum initial heat output with ones having the maximum initial heat output can be seen in Fig. 9, which shows the variation of temperature with time for this case. From Fig. 9, it is clear that the temperature at the top, bottom and in the center of the container reaches the solidus of HDSM. The coolest temperatures, which only just reach the solidus occur at the top of the container. In practice, this would not present a problem since heat from an overlying container would raise this temperature significantly, giving a good melt zone in the annuli.

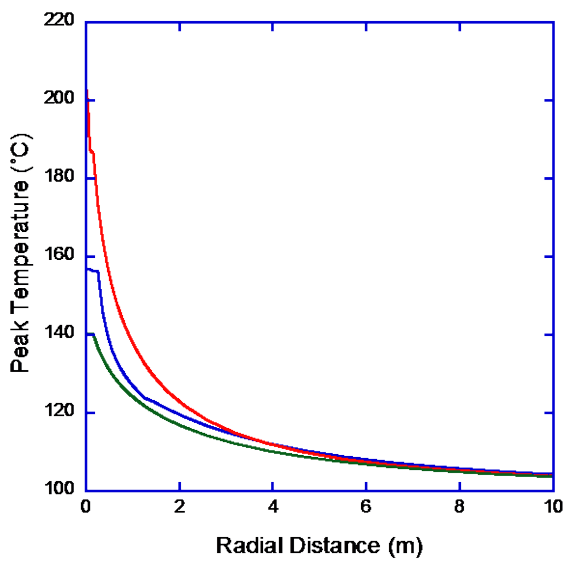

Fig. 7. Peak temperature along a horizontal radius for the "triples" concept using minimum heat output for Cs (see text). Key: red line: midway up container;blue: bottom of container;green: top of container.

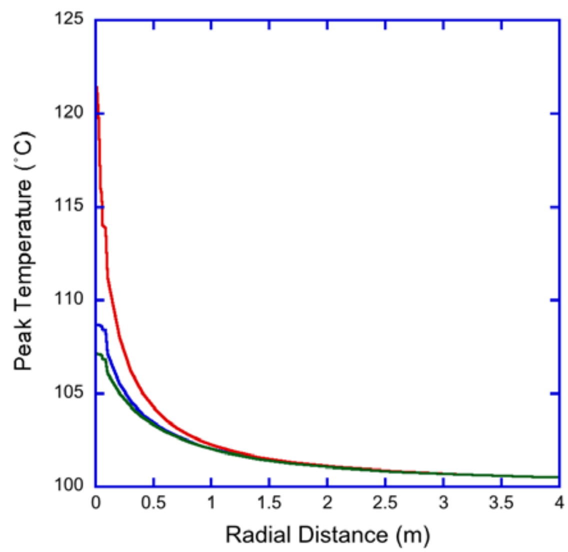

Fig. 8. Peak temperature versus radial distance for baseline concept using minimum heat output (see text) for Cs. Key as in Fig. 7

Consideration of the disposal of a single container of capsules is useful for establishing a base case from a modeling perspective but is unlikely to be of much practical use due to the low disposal efficiency (it is highly unlikely that a single borehole would be used for the disposal of such a small quantity of waste material). A more realistic scenario would involve the disposal of a stack of such containers, perhaps containing the entire capsule inventory. It is useful then to model a stack of containers. We have opted to model a stack of 10 “triple” containers which considers the simultaneous disposal of 300 Cs-containing capsules. We have taken the worst case scenario (from a heat output point of view with the intended use of HDSM in mind) and considered all 300 capsules at the low end of the predicted heat output distribution. Fig. 10 shows the variation of temperature with time for this particular case. The plot shows that the maximum temperatures are attained at longer post disposal times and that these maxima are around 10 degrees higher than for the case of a single container having the same heat output density (cf. Figs 10 and 5). This may at first appear to rule out the use of HDSM as the SSM since only the centre of the stack exceeds the solidus of the alloy. However, as can be

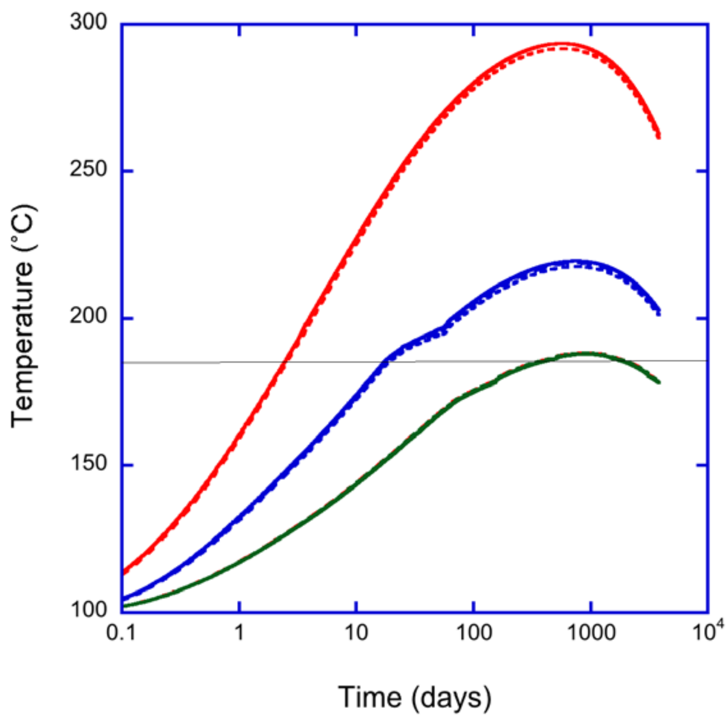

Fig. 9. Temperature versus time plot for “triples” concept using maximum heat output for Cs (see text). (Key: as for Fig. 6.)

seen by comparing Fig. 5 and Fig. 9, the difference between the maximum and minimum heat outputs for the capsules is more than double and is likely to have a similar effect on the temperature rise. In a practical disposal scenario, the capsules within a container would contain a mixture of heat outputs calculated to push the temperature rise above the HDSM solidus. Furthermore, a mixture of Cs and Sr capsules could be considered either within a single container or by having alternate containers of 100% Cs or 100% Sr. The heat density is the important factor, not the length of the column of containers. This is important to bear in mind if the entire inventory were to be disposed in a single hole of these dimensions; the additional temperature rise over and above a partial disposal would not be too significant (for the same heat density). Figs. 11-12 show that the thermal footprint from a 10 container disposal scenario is not too large; the maximum temperature rise is only a few degrees at a radial distance of only 25 m from the borehole axis while the temperature differential across the vertical height of the stack of containers is only

11 °C – an important fact when considering the possibility of convection currents within an (unfilled) borehole annulus.

Convective transport calculations

Convective transport within the borehole annuli is only a serious problem for holes with unfilled annuli. In our concept, the annuli are filled with a HDSM, removing this possibility. However, there remains the potential for upward transport of an escaped radionuclide via convection in the fluids within the granite rock.

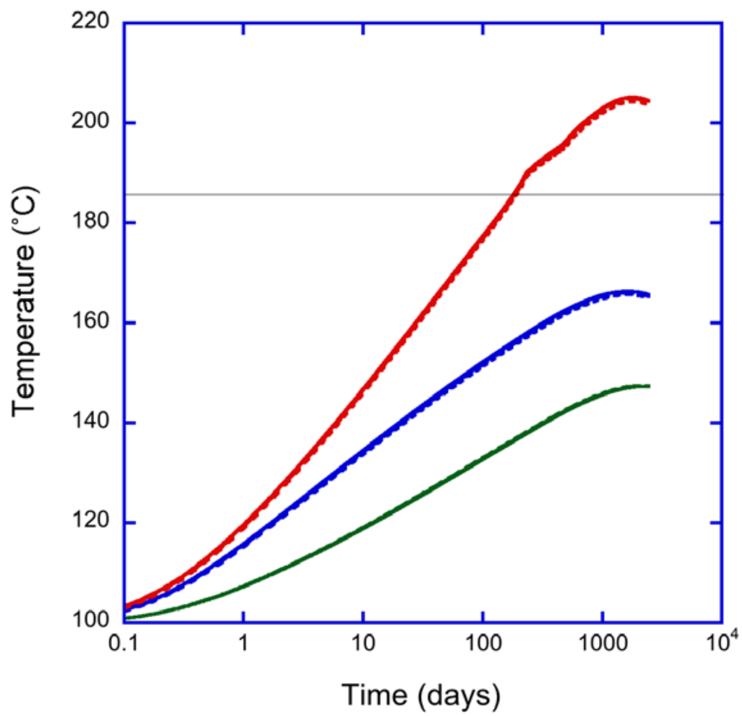

Fig. 10. Temperature versus time plot for 10 "triples" containers using minimum heat output (see text) for Cs. (Key: as for Fig. 5.)

To obtain an estimate of the vertical distance travelled by this mechanism, we have solved Darcy's law within the Boussinesq approximation:

$$\frac{\eta}{k}u = -\nabla\left(p_0 + p\right) + \rho_0 g\beta\hat{\mathbf{k}}\left(T - T_0\right) - \rho_0 g\hat{\mathbf{k}} \qquad \text{(Eq. 3)}$$

where u is the velocity of a fluid element, η the fluid viscosity, k the permeability, p is pressure, p_0 the hydrostatic pressure, β the thermal expansion coefficient, g the gravitational acceleration, ρ_0 is the density at ambient temperature $T = T_0$ and $\hat{\mathbf{k}}$ is a unit vector in the vertical (z) direction. The temperature rise can be approximated as that due to a point source of heat via

$$T - T_0 = \frac{q}{4\pi\kappa r} \qquad \text{(Eq. 4)}$$

where κ is the thermal diffusivity and q is the heat output per unit time per unit volume. Finally, we assume incompressibility:

$$\nabla \cdot \mathbf{u} = 0 \quad \text{(Eq. 5)}$$

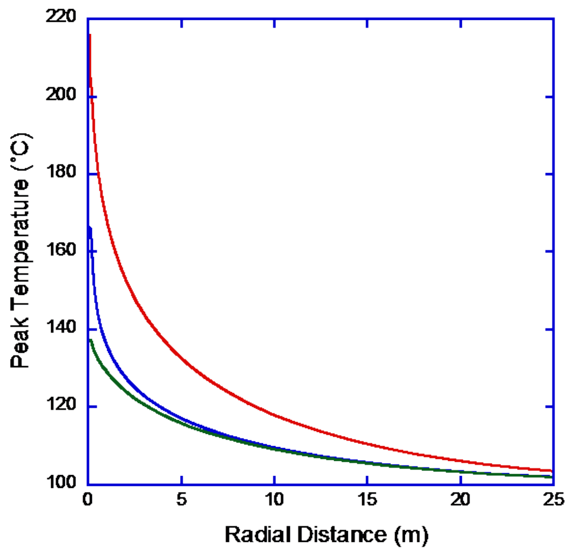

Fig. 11. Peak temperature along a horizontal radius for the baseline concept using minimum heat output for Cs (see text). Key: as in Fig.7.

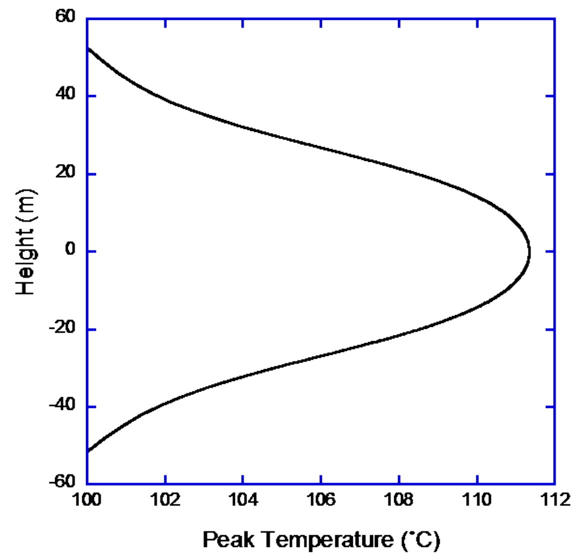

Fig. 12. Peak temperature at the surface of a stack of 10 "triples" containers calculated as a function of vertical distance relative to midpoint of the stack using minimum heat output for Cs (see text).

Solving Eq. 3 together with Eqs. 4-5 results in an implicit equation (Eq. 6) for the vertical distance travelled by a particle in terms of the elapsed time [9].

$$t = \frac{a^2}{C}\left[\frac{1}{2}\sinh\lambda\cosh^3\lambda + \frac{3}{4}\sinh\lambda\cosh\lambda + \frac{3}{4}\lambda\right] \quad \text{(Eq. 6)}$$

For a given value of time and specified constants a and C, Eq. 6 was solved using Matlab™ for the variable λ. This value of λ is then used to obtain the value of z:

$$z = a\cosh\lambda\sinh\lambda \quad \text{(Eq. 7)}$$

The constant $C = \frac{\psi\beta q}{4\pi\kappa}$, where ψ is the hydraulic conductivity and κ is the thermal diffusivity of the porous medium. The constant a is the radial position of the particle when z = 0. In this work we took a value for a of 0.16 m which corresponds to the interface between the borehole and the rock. We also explored values 10 and 100 times this value. The parameter with the greatest degree of uncertainty is the hydraulic conductivity. We have used a value of 10^{-11} for this quantity, based on values appropriate to granite and water at 100 °C.

Fig. 13 shows convection results for both a single container and a stack of 10 containers, each with 30 capsules of caesium chloride with the minimum heat output. Results are also shown for three different radial starting points as discussed earlier. Evidently the vertical migration due to convection increases with time but diminishes with radial distance. The most striking feature of these calculations is the magnitude of the distance travelled. For example, in 100,000 years, a particle starting at the rock wall would move a distance of only 5.7 m. Since these calculations represent an upper bound, the actual distance can be expected to be less than this value. We carried out a sensitivity analysis to the hydraulic conductivity. Using values of 10^{-10}, 10^{-9} and 10^{-8} gave corresponding vertical distances of 18 m, 58 m, 184 m respectively for the 10 container example with the particle starting at a radial distance from the borehole axis of 0.16 m. Based on these calculations, convective transport through the porewaters within the granitic rock can be largely disregarded.

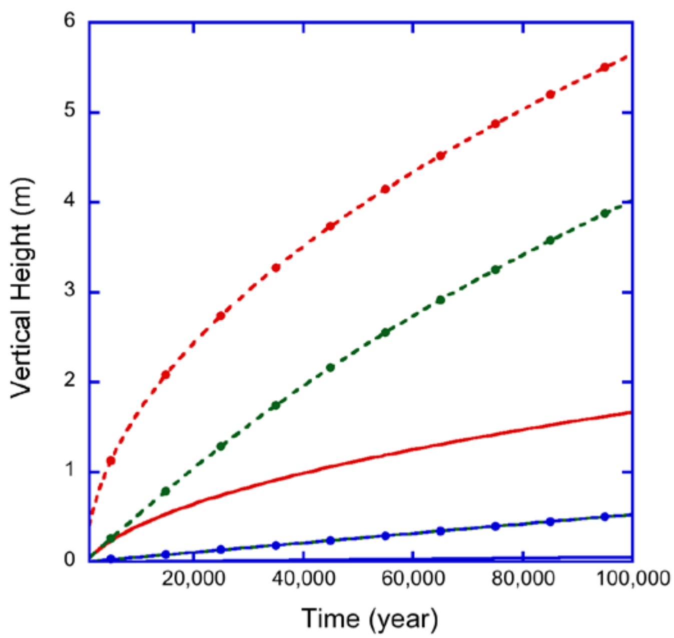

Fig. 13. Vertical distance travelled by a hypothetical particle via convective transport through the granitic rock using triples concept. Key: solid lines: 1 container; dashed lines: 10 containers; Red: *a* = 0.16 m; Green: *a* = 1.6 m; Blue: *a* = 16 m.

CONCLUSIONS

We have extended our previous baseline DBD concept for disposing of the Hanford CsCl capsules in a single borehole. Our new concept involves placing 30 capsules into each container arranged as 5 rows of 3 end-to-end aligned capsule pairs. A disposal container of this size requires a borehole with a diameter of only 31.1 cm (12.25 inch) (compared to the 21.6 cm (8.5 in) needed for our baseline concept). With more capsules per container, a much shorter disposal zone (DZ) is required which could provide either a greater geological barrier or allow a shallower hole with likely cost savings.

Focusing on the disposal of Cs, our modeling work has demonstrated the feasibility of using a HDSM as a sealing and support matrix – our preferred choice of annulus filler. Taking the extreme case of a container with capsules all having the maximum initial heat output, the results show that enough decay heat could be released to melt the HDSM surrounding a single container. For the minimum heat output example, the HDSM is only melted around the center (hottest part) of the container. Increasing the disposal volume to a stack of 10 containers, all with the minimum heat output raises the maximum temperatures above those for a single container, but not substantially so. The disposal zone for the stack still possesses a small thermal footprint. From the perspective of the HDSM, a greater temperature rise at the top and bottom of the stack, sufficient to melt the shot at these points could always be arranged by mixing capsules with different heat outputs within the same container or alternating higher and lower heat generating containers in the stack, possibly mixing in some strontium capsules to increase heat density.

By considering the possibility of upward transport of an escaped radionuclide via the fluids in the granitic host rock, we have determined that this form of transport would be insignificant for a disposal using our triples concept.

It would appear that disposal of the Hanford capsules in a 12.25 in diameter borehole – well within current drilling capabilities for a vertical hole drilled to 4-5 km depth is a realistic and appealing possibility, particularly when used with the HDSM filling the borehole annulus.

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