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SUBJECT:

THERMAL CONSIDERATIONS FOR EMPLACED WASTE PACKAGES.  
AN EA REVIEW DRAFT WORKING PAPER

In the course of our review of the DOE Environmental Assessment documents for potential salt sites, we undertook a review of heat transfer from waste packages to the host salt. The objective was to make an independent assessment of the temperature profiles reported in previous studies. Through this assessment, thermal properties and especially thermal conductivities, were identified as sensitive parameters.

Overall, we conclude that excessive thermal loadings may be imposed on the host rock by the Alternate II designs and that liquid water is unlikely to be present in the very near field for at least a few hundred years after emplacement.

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We, therefore, recommend that a definitive review of temperature and moisture profiles in the very near field be made using three dimensional models.

We would appreciate receiving comments on the enclosure.

**ORIGINAL SIGNED BY**

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Enclosure:  
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Enclosure 1

Thermal Considerations for Emplaced Waste Packages

An EA Review Draft Working Paper

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Enclosure 1  
Thermal Considerations for Emplaced Waste Packages  
An EA Review Draft Working Paper

Introduction

Emplacement of high-level waste (HLW) packages in geologic repositories introduces a transient heat source which produces a long-lasting general elevation of temperature within the waste packages, the controlled area defined in 10 CFR Part 60 (Reference 1), and the geologic setting outside the controlled area. The design of the waste packages and the repository must be such as to prevent these temperatures from exceeding certain limits which are based mainly on the properties of the materials of construction.

Much work has been done to predict temperature profiles for particular designs. Because of the complex geometry of the repository, these predictions have been obtained through use of mathematical models and computer techniques of varying degrees of sophistication. While the results appear plausible, it is important to have an independent way of assessing and confirming them. One concern is that models generally require simplifications and assumptions and so may not represent the real situation adequately. A second concern is that calculations that can be made using simpler models indicate a potential for substantial and possibly unacceptable temperature rises in and near the waste packages. This memorandum presents some of these calculations and identifies information needed to assess DOE conclusions on temperature profiles and consequent assertions as to waste package life.

Scope

The analysis is basically in terms of a salt repository, but some applications to a basalt repository are included. The analysis uses approximate methods to estimate temperatures significant to waste package and repository designs. The focus is on independently assessing the validity of previously calculated tem-

perature profiles, identifying sensitive parameters, and providing guidance for the engineering design efforts.

The logic of this approach is to let DOE remain responsible for the definitive assessment of temperature profiles via models, simulations, and field tests while probing the proposed repository design by means of selected limiting cases. If different conclusions are reached, the probing offers insight into the origins of the differences.

### Waste Package Design Parameters

Three types of HLWs require disposal: commercial high-level waste (CHLW), defense high-level waste (DHLW), and spent fuel (SF2). The environmental assessment (EA) documents released by DOE for salt sites (Reference 2) show that Alternate II as described in a 1983 Battelle report (Reference 3) is the currently favored design option for at least CHLW and SF2. Selected design data are given in Table 1. The cylindrical packages are assumed to be emplaced in vertical boreholes in the tunnel floors in the repository.

### Basic Heat Conduction Equations

The first few cases we shall examine deal with transfer of heat by conduction only, for which the temperature distributions are obtained as the solutions of a second order differential equation which in rectangular coordinates is:

$$1a. \quad c\rho \frac{\partial T}{\partial t} = q_v + \sum_i \frac{\partial}{\partial x_i} \left[ k \frac{\partial T}{\partial x_i} \right], \quad i = 1, 2, 3$$

and in cylindrical coordinates is:

$$1b. \quad c\rho \frac{\partial T}{\partial t} = q_v + \frac{1}{r} \frac{\partial}{\partial r} \left[ rk \frac{\partial T}{\partial r} \right] + \frac{1}{r^2} \frac{\partial}{\partial \theta} \left[ k \frac{\partial T}{\partial \theta} \right] + \frac{\partial}{\partial z} \left[ k \frac{\partial T}{\partial z} \right]$$

In these equations,

$T$  = temperature at any point, K

$t$  = time, s

$c$  = heat capacity, J/kg-K

$\rho$  = density, kg/m<sup>3</sup>

Table 1. Selected Waste Package Design Data  
Basis: Alternate II, ONWI-438, Battelle, April 1983

Dimensions	CHLW	DHLW	SF2/PWR
<u>Canister</u>			
Material	316 SS	304 L	316 SS
O.D., m	0.560	0.610	0.570
I.D., m	0.5346	0.591	0.5573
L, m (w pintle)	4.09	3.00	4.10
<u>Waste Form</u>			
Volume, m <sup>3</sup>	0.825	0.626	-
L, m	3.675	2.282	3.85
<u>Overpack</u>			
Material	Carbon steel	Carbon steel	Carbon steel
O.D., m	0.890	0.840	0.835
I.D., m	0.590	0.632	0.595
L, m (ex pintle)	4.57	3.38	4.48
Corrosion allowance, cm	5.00	0.90	2.50
<u>Borehole</u>			
I.D., m	0.940	0.890	0.890
L, m	6.3	4.5	5.9
Package pitch, m	10	2.39, square	10.4
<u>Tunnel</u>			
Height, m	7.2	6.1	7.4
Width, m	4.0	5.0	4.0
Spacing, m	31.6	25.8	28.3
<u>Other Parameters</u>			
<u>Waste Form</u>			
Weight, kg			
Total	3,425	1,940	8,590
Waste	2,560	1,470	4,610 kg U
Canister	865	470	510
Overpack	13,425	7,360	10,410
Heat Load, kW	9.500	0.423	5.500
W/kg waste	3.711	0.2878	1.193 kg U
Radiation, rem/hr			
Overpack surface			
Gamma	95	130	85.1
Neutron	4.96	NA	2.23
<u>Repository</u>			
Plan area, m <sup>2</sup> /pkg	316	30.83	294.3
Height, m/pkg	4.57	3.38	4.48
Volume, m <sup>3</sup> /pkg	1,444	104.2	1,319
Area heat release, W/m <sup>2</sup>	30.06	13.72	18.69

$k$  = thermal conductivity, W/mK  
 $q_v$  = volumetric heat release, W/m<sup>3</sup>  
 $x_i$  = linear distance from some origin, m  
 $r$  = radial distance from the waste package axial centerline, m  
 $z$  = axial distance from the origin, m  
 and  $\theta$  = angular displacement from the r-axis, radians.

We may also define  $\alpha^2 = k/c_p$ , m<sup>2</sup>/s, as the thermal diffusivity of the conducting medium.

What these equations express is the time rate of increase in the temperature in a medium (the term on the left side of the equation) due to accumulation of heat from an internal source,  $q_v$ , less the net heat loss by conduction. To identify NRC concerns relevant to the waste packages, we shall make use of selected special cases.

#### Heat Conduction in the Host Rock

Case 1. Zero conductivity outside the waste package, constant heat source within the package.

This case is of primary interest because without conduction of heat into the host rock the temperature within the waste packages rises without limit. An equivalent description of this case is that for a waste package with an adiabatic boundary, i.e., for a heavily insulated package. Radial models in effect use this assumption for the axial boundaries. In the actual situation, the temperature rise within the waste package due to a constant internal heat source reflects the balance between the storage capacity (i.e., heat capacity) of the package and the ability of the salt to conduct heat away from the package. An appreciation of how rapidly this temperature can rise is needed in assessing both design and operation. If the heat source is considered to be uniformly distributed within the waste package, the temperature gradient in all three directions is everywhere zero, and the equation 1b reduces to:

$$2. \quad c_p \frac{dT}{dt} = q_v,$$



which, by assuming the density and the specific heat are constant or may be represented by average values, may be integrated directly to:

$$3. \quad \frac{\Delta T}{\Delta t} = \frac{q_v}{c_p} = \frac{Q}{wc},$$

where

$Q$  = total heat release within the package, W  
and  $w$  = mass of the waste package, kg.

Using the design values in Table 1, we find the following temperature increase rates for the waste plus the canister:

$$\text{For CHLW: } \frac{\Delta T}{\Delta t} = \frac{(9500 \text{ W})(3600)}{(2560 \text{ kg})(750) + (865 \text{ kg})(502)} = 14.5 \text{ K/hr}$$

$$\text{For SF2: } \frac{\Delta T}{\Delta t} = \frac{(5500 \text{ W})(3600)}{(8080 \text{ kg})(264) + (510 \text{ kg})(750)} = 7.87 \text{ K/hr}$$

$$\text{For DHLW: } \frac{\Delta T}{\Delta t} = \frac{(423 \text{ W})(3600)}{(1470 \text{ kg})(750) + (1475 \text{ kg})(502)} = 0.113 \text{ K/hr}$$

These rates of temperature rise would occur over the short term if there were no heat losses from the waste packages and if all the heat generated within them was uniformly distributed over the mass of the waste form and the canister, with allowance for differences in specific heats. In 24 hours, the temperature of the CHLW package could increase 350°C, while the SF2 package could experience a similar rise in 44 hours. The same rise would require about four months for the DHLW package.

It is clear that the previous temperature history of the packages must also be considered. The CHLW waste form may be produced at an initial temperature of as much as 1000°C. Presumably this is true also for the DHLW waste form. The fuel rods, on the other hand, have been exposed to at least 600°C in the reactor. All waste forms will be stored for some time prior to shipment to the repository, and while in storage all must be cooled continuously. Without going into a detailed review of the cooling options during initial storage, shipment to the repository, and interim storage at the repository, it appears likely that both

CHLW and SF2 may be emplaced at 100-150°C, and thus may be at 450-500°C within one or two days. Table 2 presents operating temperature limits for various materials of interest in HLW disposal. The adequacy of the cooling process for emplaced packages thus merits a critical review. This review should cover temperature profiles within the packages as well as in the host rock.

## Case 2. Finite conductivity in the host rock, constant heat source

### A. Conduction within tunnels

Consider the cylindrical annulus of salt around a waste package. We assume the borehole is backfilled with crushed salt which rapidly (by creep) takes on the properties of solid salt so there is no air gap. To simplify equation 1b, we neglect axial conduction, and assume radial symmetry (at least for the near term):

$$4. \quad c_p \frac{\partial T}{\partial t} = \frac{1}{r} \frac{\partial}{\partial r} (rk \frac{\partial T}{\partial r})$$

The solution of this equation involves Bessel functions, but we may visualize a limiting case to gain further insight into the nature of the heat transfer process. While the radionuclide heat source does decay with time as reported in Table 3, let us suppose that the time interval for decay is long compared to that for heat conduction. A quasi-steady state therefore will prevail for some period of time, so that the equation 4 further reduces to:

$$5. \quad 0 = \frac{\partial}{\partial r} (rk \frac{\partial T}{\partial r}), \text{ which means that}$$

$$6. \quad rk (dT/dr) = c_1, \text{ a constant}$$

The value of  $c_1$  is obtained from the boundary condition that at steady state:

$$7. \quad Q = -k S (dT/dr),$$

Table 2. Operating Temperature Limits for Various Materials  
in High Level Waste Disposal Service<sup>a</sup>

Material	Temperature Limit, °C	Reason
Borosilicate glass	500	Devitrification
Fuel rods		
Spent fuel pins	300	Stress rupture
Zircalloy cladding	375	Corrosion
	200	Reactivity to oxygen
Water	374	Critical temperature
Salt (natural deposits)	250	Decrepitation
Basalt	200	Thermal cracking
Lead	328	Melting
Carbon steel	430	Oxidation in air; loss of strength by creep
Ticode-12	250	Corrosion
Bentonite	150	Hydrothermal <sub>b</sub> alteration <sub>b</sub>

<sup>a</sup>Reference 4, Table 4.1, page 83.

<sup>b</sup>Reference 13

where  $S$  is the cylindrical surface of the overpack, which in turn is given by  $2\pi r_1 L$ , where  $r_1$  is the outer radius of the overpack and  $L$  is the length of the overpack. Hence,

$$8. \quad c_1 = -Q/(2\pi L)$$

Integrating from  $r_1$  to  $r_2$  (an arbitrary outer radius of the salt annulus), assuming  $k$  is constant or that an average value applies:

$$9. \quad k (T_1 - T_2) = \frac{Q}{2\pi L} \ln (r_2/r_1)$$

where  $k_m$  = mean thermal conductivity in the temperature range  $T_1$  to  $T_2$ .

Now in choosing Alternate II, DOE increased the heat generation within the CHLW packages from 2210 W to 9500 W. As compensation, DOE also increased the package spacing within tunnels from 2.48 to 10 m. From the data in Table 1,

$$10. \quad k (T_1 - T_2) = \frac{9500}{2\pi(4.57)} \ln (5/0.445) \\ = 800.4$$

For an average  $k_m$  of 4.1 W/m-K, and an ambient salt temperature  $T_2 = 33^\circ\text{C}$ , the temperature at the surface of the CHLW overpack will be  $228^\circ\text{C}$ . As a check, linear interpolation between the two boundary temperatures yields  $k_m = (5.3 + 2.8)/2$  or 4.05. This is actually somewhat high because  $k$  vs temperature is not linear and because the cylindrical geometry requires a logarithmic interpolation. It is, however, close enough for the purposes of this analysis. The calculation clearly shows how critical the value of  $k$  is. According to the literature<sup>a</sup>,  $k$  for crushed salt is only one-tenth that of solid salt. If the value of  $k$  is only half that used above, the surface temperature for the CHLW package would be  $423^\circ\text{C}$ .

We also need to check the time element: how quickly does the annulus of salt heat up? Proceeding as before with equation 3, using typical values for heat capacity and a density, the mass of the annulus is:

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<sup>a</sup>Reference 3, page 335; Reference 4, Page 30.

$$\begin{aligned}
 11. \quad W_1 &= (\pi/4)[(10.0)^2 - (0.890)^2] (4.57)(2170 \text{ kg/m}^3) \\
 &= 7.73\text{E}+05^b \text{ kg},
 \end{aligned}$$

and the rate of temperature rise is:

$$\begin{aligned}
 12. \quad \frac{\Delta T}{\Delta t} &= (9500)(3600)/(W_1)(850 \text{ J/kg-K}) \\
 &= 0.0521 \text{ K/hr.}
 \end{aligned}$$

Thus, in 280 days, the temperature throughout the annulus might have risen 350°C. Actually, the heat conduction along the tunnels will begin to decrease as soon as heat from adjacent packages reaches the interface between them at 5 meters from their axial centerlines. The vertical planes at these locations become adiabatic boundaries at which the temperatures will rise, thus reducing the temperature gradient for heat transfer in this direction. The salt within the tunnels then becomes a storage medium for a portion of the decay heat.

A similar result is obtained for the SF2 packages, for which Q is 5500 W/package.

$$\begin{aligned}
 13. \quad k (T_1 - T_2) &= \frac{5500}{2\pi(4.48)} \ln (5.2/0.4175) \\
 &= 492.8
 \end{aligned}$$

With  $k_m = 4.4$ ,  $T_1 = 144^\circ\text{C}$ ; for  $T_1$  to exceed  $400^\circ\text{C}$ , the value of  $k_m$  would have to be less than 30% of the value used here. The mass of the salt annulus is:

$$\begin{aligned}
 14. \quad W_2 &= (\pi/4) [(10.4)^2 - (0.835)^2] (4.48) (2170) \\
 &= 8.20\text{E}+05 \text{ kg},
 \end{aligned}$$

and the rate of temperature rise is:

$$\begin{aligned}
 15. \quad \frac{\Delta T}{\Delta t} &= (5500)(3600)/(W_2)(850) \\
 &= 0.0284 \text{ K/hr.}
 \end{aligned}$$

---

<sup>b</sup>Fortran exponential notation.

A 350°C rise thus requires about 1.4 years.

The result for DHLW is somewhat suprising, because the heat generation is so much less: 423 W/package.

$$\begin{aligned} 16. \quad k_m(T_1 - T_2) &= \frac{423}{2 \pi (3.38)} \ln (1.95/0.42) \\ &= 20.83 \end{aligned}$$

With  $k_m = 5.2$ , the surface temperature of the overpack is 37°C; at one-tenth this value,  $T_1$  would still be only 73°C. This means that if a steady state condition were reached, the heat generation within a DHLW package could be dissipated in the salt with only a minor elevation in package temperature. However, the mass of the salt annulus in this case is much less:

$$\begin{aligned} 17. \quad W_3 &= (\pi/4) [(2.39)^2 - (0.84)^2] (3.38) (2170) \\ &= 2.88E+04 \text{ kg,} \end{aligned}$$

and the rate of temperature rise is

$$\begin{aligned} 18. \quad \frac{\Delta T}{\Delta t} &= (423) (3600)/(W_3) (850) \\ &= 0.0621 \text{ K/hr.} \end{aligned}$$

Here, a 350°C rise requires 235 days. We interpret the foregoing results to mean that, although only a small temperature rise is required to conduct heat out of the waste package, in about 8 months the salt between packages within tunnels will have heated enough to substantially reduce further conduction in this direction. Meanwhile, the temperature within the packages will gradually rise to provide the necessary differential to conduct the heat generated in one of the other two directions.

We conclude that within a small fraction of the time period during which the heat generation rate within the packages remains essentially constant at emplacement levels, heat conduction along the tunnels becomes a minor fraction of the total heat release from the packages.

## B. Conduction between tunnels

To examine this case, we consider the simplified model for the waste packages to be rectangular blocks whose dimensions are the package pitch within tunnels x tunnel spacing x overpack height. Half the heat generated is conducted toward each of the two adjacent tunnels, since we are still neglecting axial conduction. The calculations are summarized in Table 4.

Table 4. Heat Conduction Between Tunnels

Waste Package	CHLW	SF2	DHLW
Heat flux, W/m <sup>2</sup>	103.9	59.0	52.4
Salt mass, Mg	1557	226	1431
Temperature rise, K/hr	0.0128	0.0079	0.0081
Time for 350°C rise, yrs	3.1	5.0	4.9

The calculation for DHLW takes into account there are two rows of packages in each tunnel. We conclude that within 3 to 5 years after emplacement the main path available for heat conduction is in the z, or vertical direction. Thus, all of the salt in the plane of the repository becomes a storage medium for heat.

## C. Conduction in the vertical direction

The simplified source model becomes a rectangular plane whose dimensions are the package pitch within the tunnel x the tunnel spacing, with half the heat flowing upwards and the other half downwards. In the case of the DHLW, one-half the tunnel spacing is used because there are two rows of packages within each tunnel. We first show that it is reasonable to treat this case as heat conduction in a semi-infinite solid.

### 1. Steady state conduction, long-lived heat source

If the heat source remained constant sufficiently long, the upward temperature pulse would eventually reach the Earth's surface, which is assumed to remain constant at 15°C. Assuming the repository has been backfilled with salt that

then rapidly consolidates, the temperature gradient required to hold temperatures within the waste packages constant may be estimated as follows for the CHLW package:

$$\begin{aligned}
 19. \quad \frac{dT}{dz} &= - Q/kA \\
 &= - 4750/(4.0)[(10)(31.6)] \\
 &= - 3.76 \text{ K/m}
 \end{aligned}$$

An implicit assumption here is that the vertical heat flow is collimated, which might prove applicable to centrally located packages. For a repository depth of 600 m, the temperature at the upper ends of the packages is thus 2270°C, potentially. However, the quantity of heat stored in the mass of salt between the repository and the Earth's surface is:

$$\begin{aligned}
 20. \quad Q_T &= (316)(600)(2170 \text{ kg/m}^3)(850 \text{ J/kg-K})(2270 + 15)/2 \\
 &= 4.09\text{E}+14 \text{ J},
 \end{aligned}$$

and the time required to generate this amount of heat is:

$$21. \quad t = Q_T/4750, \text{ or } 2674 \text{ years.}$$

It is clear that the heat source will decay long before the temperature pulse reaches the Earth's surface, and therefore it is valid to consider the salt mass as semi-infinite in the vertical z-direction. However, this calculation also shows that the role of the host rock is initially that of a temporary storage medium for decay heat as distinguished from that of a medium for conducting heat for dissipation to the atmosphere.

## 2. Transient conduction, constant heat source

From Carslaw and Jaeger (Reference 5), the temperature distribution in a semi-infinite solid subjected to a constant heat flux at the plane  $z = 0$  with the initial temperature everywhere in the solid at 0°C is:

$$22. \quad T = \frac{2 F_0 \sqrt{\alpha^2 t}}{k} \text{ierfc}(Z/2\sqrt{\alpha^2 t})$$



where  $F_0$  = Heat flux,  $W/m^2$

and  $ierfc$  is the integral of the complement of the error function.

Some of the symbols have been changed from those in C&J for consistency with those used earlier this memorandum. At  $z = 0$ , the value of the  $ierfc$  term is  $1/\sqrt{\pi}$ . Because of the dependence of the thermal parameters  $\alpha^2$  and  $k$  on temperature, and because equation 22 was obtained for constant thermal properties, suitable average values are needed. Arithmetic averages were used based on the background temperature of  $33^\circ C$  and the calculated package surface temperature, the latter being obtained by a trial-and-error procedure. Calculations are summarized in Table 5.

Table 5. Heat Conduction for a Semi-Infinite Solid

Waste package	CHLW	SF2	DHLW
$Q$ , W	4750	2750	212
$F_0$ , $W/m^2$	15.03	9.34	6.89
$T$ @ $z = 0$			
At 1 year	64	52	47
At 10 years	138	96	89
$z$ @ 10 years, m			
$T = 100^\circ C$	13.4	-	-
$T = 66^\circ C$	30.1	18.9	10.8
$T = 43^\circ C$	55.9	46.5	39.5

The temperatures shown include the background value of  $33^\circ C$ . Note that all these temperatures are inversely proportional to  $\sqrt{k}$ , so that if  $k$  is only one-fourth as large, the temperatures at  $z = 0$  in 10 years would be 243, 158, and  $145^\circ C$  for the CHLW, SF2, and DHLW packages respectively. To follow this thought, the thermal conductivity of water over the range  $0-327^\circ C$  goes from  $0.56 W/m-K$  to  $0.68$  at  $149^\circ C$  and then drops to  $0.48 W/m-K$ . Shale and sandstone appear to have  $k$  values at  $20^\circ C$  in the range  $1.4-2.8 W/mK$ . The  $k$  values for irradiated salt appear to lie below those for unirradiated salt. Thus, there is reason to believe actual thermal conductivities could be significantly less than those for pure halite.

To provide some perspective, the vapor pressure of pure water rises to 10 atmospheres at about  $180^\circ C$ . The presence of dissolved salts ( $NaCl$ ,  $KCl$ ,  $CaCl_2$ )

could depress this to about 3 atmospheres. The tensile strength of salt, either bedded or domal, is 1-2 MPa (10-20 atmospheres) but decreases with increasing temperature, hence temperatures in the salt above 180°C appear excessively high. To provide a factor of safety of even two suggests temperatures anywhere in the salt should not exceed 150°C. At this temperature, the vapor pressure of pure water is about 5 atmospheres. Further study might lower this suggested temperature even further.

Equation 22 can also be used to estimate penetration of the temperature pulse into the host rock by solving for the value of  $z$  that yields particular temperatures. The results are also shown in Table 5 for three isotherms at 10 years. These results indicate temperature increases within the suggested limit, especially when one considers the heat source has been decaying for ten years. Whether this is so merits a more definitive study using better thermal property data. However, these results suggest that more attention should be focussed on the response of the salt in the plane of the repository between waste packages. Simple radial geometry appears insufficient. There will be radial conduction from each waste package to adiabatic boundaries which form a rectangular block of salt plus axial conduction into rectangular blocks of salt lying above and below the packages. Numerical methods of analysis will probably be required.

### 3. Transient conduction downwards

It appears that previous models have treated the host rock as an infinite sink, so that there is vertical symmetry in the heat conduction process. There is, however, an omission in this model. It is recognized that geothermal heat results in a thermal gradient up toward the surface of the Earth of about  $0.030 \text{ K/m}^a$ . This is equivalent to a flux on the order of  $(4 \text{ W/mK})(0.030 \text{ K/m})$  or  $0.120 \text{ W/m}^2$ . It has apparently been assumed that this has a negligible effect on the repository compared to the design fluxes from the repository of  $10\text{-}15 \text{ W/m}^2$ .

It must be remembered, however, that heat will flow only under a temperature gradient. Consequently, the small geothermal gradient observed near the surface of the Earth could really be a result of heat-generating processes deep within the Earth.

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<sup>a</sup>Reference 6, Figure 2-31, page 165, shows  $0.045 \text{ K/km}$  for Hanford basalt.

Assuming the near surface thermal gradient remains constant with depth, temperatures of 500°C would be found 16 km below the surface. Heat added to the rock by the repository would increase the rock temperature, temporarily decreasing the gradient from below the repository. The long term response at depth would be an increase in temperature, restoring enough of a gradient to resume conduction of the 0.120 W/m<sup>2</sup>. An alternative is to absorb the unconducted heat in phase changes, as melting of additional amounts of rock.

We think this scenario means all the heat conducted initially downward from the repository will eventually return to the repository and ultimately be dissipated at the Earth's surface. The action is something like that of trying to roll a ball uphill. As the ball rolls uphill, its initial velocity decreases until finally it reverses direction and rolls downhill under the influence of gravity. If the repository temperature peaks before the heat being conducted downward encounters comparable temperatures, then the effect of the return wave may be merely to prolong the time the repository remains at elevated temperatures, although these will be below the initial peak. Also, not all the heat will return to the repository, since some of it will be conducted laterally beyond the perimeter of the repository.

#### 4. Conduction versus Storage

In summary, the foregoing sections indicate the following sequence of events. Upon emplacement, the temperature of the waste packages will be above the temperature of the ambient salt. Heat will flow from the packages radially and axially, but the packages will continue to heat up because more heat is being generated than can be conducted away by the initial small temperature gradients. Relatively soon, the rate of heat conduction along tunnels will diminish because of the presence of other packages in those tunnels. Somewhat later, heat conduction between tunnels will also diminish for a similar reason. At some time, a maximum temperature will be reached within the packages because the radio-nuclides will have decayed sufficiently. Because the decay is slow, we may consider that at this time a quasi-steady state has been reached such that a constant gradient exists at the surface of the slab source representing the waste package array. The heat generated to this time,  $Q_t$ , will be divided between that going toward elevation of the temperature of the slab source and

that conducted into a block of host salt of height  $z$ . For a CHLW package, the heat in joules is:

$$\begin{aligned}
 23. \quad Q_t &= 10 \text{ m} \times 31.6 \text{ m} \times (4.57 \text{ m}/2)(2170 \text{ kg}/\text{m}^3) \times (850 \text{ J}/\text{kg}\cdot\text{K}) \times (T - 33) \\
 &\quad + (10 \text{ m} \times 31.6 \text{ m} \times z) \times 2170 \times 850 \times (T - 33)/2 \\
 &= (1.332\text{E}+09)(T - 33) + (2.914\text{E}+08)(T - 33)(z)
 \end{aligned}$$

Half of the heat leaving the packages is conducted upwards, and the other half downwards.

In  $t$  years after emplacement, the heat generation rate  $P$  will have decayed to some lower percentage of the emplacement value.<sup>a</sup> The total heat release may be estimated from an arithmetic value,  $P_{av}$ .

$$24. \quad Q_t = (9500 \text{ W})(P_{av}/100)(31.56\text{E}+06)(t).$$

The heat conducted into the host rock will have been conducted under an average of the initial gradient and final gradients:

$$\begin{aligned}
 25. \quad Q_c &= (4 \text{ W}/\text{m}\cdot\text{K})(3160 \text{ m}^2) \frac{(T - 33)}{2z} (31.56\text{E}+06)(t) \\
 &= (2.914\text{E}+08)(T - 33)(z)
 \end{aligned}$$

$$\text{Hence} \quad z = 26.16 \text{ } t^{\frac{1}{2}}.$$

At 10 years after emplacement,  $P$  will be 70%, and  $P_{av}$  will be 85%;  $Q_t$  then is  $(9500)(0.85)(31.56\text{E}+06)(10)$  or  $2.548\text{E}+12$  joules. The value of  $z$  is 82.7 m, and  $(T - 33)$  is  $100.2^\circ\text{C}$ .  $Q_c$  is then  $2.414\text{E}+12$  joules, or 95% of the total heat generated.

Thus, the heat generated by CHLW packages will have penetrated 83 meters above and below the repository. This approximation is consistent with Table 5.

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<sup>a</sup>Reference 7, page 61.

## Heat Conduction in the Waste Packages

### Case 1. Heat transfer across assembly gaps

#### A. Overpack/borehole gap

Neglecting heat conduction out the top and bottom of the waste packages, the heat generated within each will be transferred to the borehole wall (in the absence of backfill) by the combined mechanisms of radiation, convection, and conduction across the annular air gap:

$$26. \quad Q/A = F\sigma (\epsilon T_1^4 - \alpha T_2^4) + h_m (T_1 - T_2) + \frac{k}{r_1} \frac{(T_1 - T_2)}{\ln(r_2/r_1)}$$

where  $Q$  = total heat generated, W

$A$  = area of the outer cylindrical surface of the overpack,  $m^2$

$T_1$  = temperature of the outer surface of the overpack, K

$T_2$  = temperature of the inner surface of the borehole, K

$F$  = radiation view factor between overpack and borehole

$\sigma$  = Stefan-Boltzmann constant,  $5.669E-08 \text{ W/m}^2\text{-K}^4$

$\epsilon$  = emissivity of the overpack

$\alpha$  = absorptivity of the borehole wall

$h_m$  = natural convection coefficient,  $\text{W/m}^2\text{K}$

$r_2$  = inside radius of the borehole, m

and  $r_1$  = outside radius of the overpack, m.

Equation 23 permits estimation of the total heat being transferred to the borehole wall at the instant the wall is at  $T_2$  and the overpack is at  $T_1$ . It also permits a determination of the relative importance of each of the three mechanisms for heat transfer, and an identification of important design parameters. Evaluation of the terms in this equation follows.

#### 1. Radiation

There are of course some complications in calculating radiant heat transfer, such as the distribution of wave lengths of the radiation and the angular direction of the radiation relative to the overpack surface. We are concerned

here with total emittance and absorption. The following generalizations from Rohsenow (Reference 8) apply to emissivities:

- Values in tables are often for emission normal to the emitting surface but may be used for hemispheric emissivity, especially for rough surfaces.
- Unoxidized, clean metallic surfaces have low emissivities.
- Most nonmetals and metallic oxides have emissivities above 0.8 at room temperature.
- Emissivity increases with temperature for nearly all materials.

For the overpack (oxidized, low carbon steel),  $\epsilon$  may be taken as 0.61 for temperatures in the range of 25 to 200°C, increasing to 0.85 for heavily crusted iron. For salt, using Kirchoff's Law,  $\epsilon$  may be set at 0.8. The view factor was calculated from an equation given in Rohsenow.<sup>a</sup> For the CHLW overpack,  $F$  is 0.938. It is of interest that for infinitely long concentric cylinders,  $F$  reduces to  $r_1/r_2$ , or 0.947.

## 2. Convection

The heat transfer coefficient  $h$  is for natural (free) convection. A convenient correlation is provided in Figure 13.5-2 of Reference 8 for vertical plates, which we used as an approximation for the overpack surface. In this correlation, the logarithm of the Nusselt Number is plotted against the logarithm of the product of the Grashof and Prandtl Numbers. Transport properties for air are evaluated at the film temperature, which is taken as the arithmetic average of the values of  $T_1$  and  $T_2$ . Table 6 summarizes the calculations, which permit the following observations for the range 0 to 500°C:

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<sup>a</sup>Reference 8, page 15-46, Configuration 5.

- Heat capacity for air is essentially constant.
- Viscosity of air increases by a factor of 2. Since the thermal conductivity of air also increases by about the same factor, the Prandtl Number is essentially constant. However, the Grashof Number is reduced by a factor of 4, considering only viscosity.
- Air density decreases by a factor of about 3, decreasing the Grashof Number by an additional factor of 9.
- When these effects are combined with the volumetric coefficient of expansion, the Grashof Number increases by a factor of 100. The net effect, however, is a reduction in the convection coefficient,  $h_m$ , from 4.76 to 2.51.

### 3. Conduction

The third term on the right side of equation 1 represents steady-state conduction across an annular medium of constant thermal conductivity. For  $r_2 = r_1 + \Delta r$ , the denominator is approximately equal to  $\Delta r$ .

### 4. Calculation of $T_1$

The independent variables in equation 1 are thus  $\varepsilon$ ,  $\alpha$ ,  $h_m$ ,  $k$  and  $T_2$ . We chose to set  $T_2$  arbitrarily at 150°C, moderately above the boiling point of water. Using this and the other values given above, for CHLW:

$$\begin{aligned}
 27. \quad Q/A &= 9500 / ((4.57)(\pi)(0.890)) = 743.5 \text{ W/m}^2 \\
 &= 0.938 (5.669\text{E-}08) [0.61T_1^4 - 0.80 (423.15)^4] + \\
 &\quad 3.79(T_1 - 423.15) + 1.53 (T_1 - 423.15),
 \end{aligned}$$

Table 6. Natural Convection Coefficient vs. Temperature

	Temperature, °C			
	0	100	300	500
Viscosity, $\mu$ , kg/m-s	1.71E-05	2.18E-05	2.95E-05	3.50E-05
Heat capacity, $c_p$ , J/kg-K	1004	1010	1043	(1100)
Thermal conductivity, $k$ , W/m-K	0.0244	0.0320	0.0459	(0.0574)
Density, $\rho$ , kg/m <sup>3</sup>	1.292	0.946	0.616	0.456
N(Pr)	0.703	0.686	0.669	0.687
N(Gr)	9.79E+11	2.37E+11	3.57E+10	9.83E+09
$X = N(Pr) \cdot N(Gr)$	6.89E+11	1.63E+11	2.39E+10	6.75E+09
log X	11.84	11.21	10.38	9.83
log Y	2.95	2.8	2.5	2.3
Y	891	631	316	200
$h_m = Yk/L$ , W/m <sup>2</sup> K	4.76	4.42	3.18	2.51

Notes

1. Figures in parentheses are extrapolated.
2.  $N(Pr)$  = Prandtl Number =  $c_p \mu / k$
3.  $N(Gr)$  = Grashof Number =  $g L^3 \beta (\rho/\mu)^2 (\Delta T)$
4. Y is read from Figure 13.5-2 of Reference 8
5.  $c_p$  and  $k$  data are from NUREG-3586, Reference 7, pages 23 and 7
6.  $\mu$  is from Lange, Reference 9, page 1576
7.  $\Delta T$  is set at 50 K



from which  $T_1$  is obtained as 211°C. For the parameter values chosen, therefore, a 61 K temperature difference exists across the annular air gap. A similar calculation with  $T_2 = 250^\circ\text{C}$  yielded 66°K, which is essentially the same. Hence, the air gap T is not sensitive to the temperature level for the temperatures of interest. Next, the distribution of heat flux among the three mechanisms is given in Table 7.

Table 7. Distribution of Heat Flux by Mechanism

	Borehole Wall Temperature, °C	
	150	250
Radiation	56%	62%
Convection	31%	24%
Conduction	13%	14%

Since radiation is the dominant mechanism, the sensitivity of the heat transfer to the value of the emissivity must be examined. Differentiating Equation 26 at constant flux and constant  $T_2$ , and treating  $h_m$  and  $k$  as essentially constant, we can derive the following error formula:

$$28. \quad -\frac{\Delta T_1}{\Delta \varepsilon} = \frac{T_1}{4\varepsilon + \frac{1}{F\sigma T_1^3} \left( h_m + \frac{k}{r_1 \ln(r_2/r_1)} \right)}$$

At  $T_1 = 150^\circ\text{C}$ , and  $\varepsilon = 0.61$ :

$$\Delta T_1 = -112.5(\Delta \varepsilon),$$

which shows that a 10% increase in  $\varepsilon$  (0.61 to 0.67) reduces  $T_1$ , the overpack surface temperature, by 7 K. Hence, as the surface corrodes, its emissivity will tend to increase which in turn will tend to reduce the temperature differential across the annular air gap. We conclude, however, that for the purposes of this evaluation a 50 K differential appears reasonable.

#### B. Overpack wall temperature differential

To estimate the temperature drop across the overpack wall, we use equation 9 from the previous section on heat conduction in the host rock:

$$29. \quad k_m (T_1 - T_2) = \frac{9500 \text{ W}}{2\pi (4.57)} \ln (0.445/0.295) = 136.1$$

Here,  $T_1$  is the temperature of the inner surface of the overpack and  $T_2$  is that of the outer surface. Assuming a wrought iron overpack, the value of  $k$  does not change much with temperature in the range of interest, so an average value of 52 W/m-K was used. The temperature drop across the wall is thus only about 3°C for the CHLW packages.

### C. Canister/overpack gap

At first, one might expect another 50K drop exists between the canister outer surface and the overpack inner surface. The ratio of radii here is 0.280/0.295 or 0.949 for CHLW and 0.285/0.2975 or 0.958 for SF2, so the view factors in radiation will be essentially unchanged. However, if the canister is not corroded, its emissivity may be as low as 0.06 to 0.21. Also, the radiating surface is smaller, so the heat flux at the canister surface is greater than that at the overpack surfaces.

$$30. \quad \begin{aligned} Q/A &= 9500/((4.09)\pi(0.560)) = 1320 \text{ W/m}^2 \\ &= 0.949 (5.669\text{E-}08) [\epsilon T^4 - 0.61(487.15)^4] + \\ &\quad 3.17 (T_1 - 487.15) + \frac{1}{3.13} (T_1 - 487.15) \end{aligned}$$

With  $\epsilon$  at 0.21,  $T_1$ , the canister surface temperature, is 384°C; with  $\epsilon$  at 0.61,  $T_1$  is 269°C. Thus, the drop across the air gap between the canister and the overpack could be in the range 55 to 170°C depending on the emissivity of the canister metal.

Similar results are obtained for SF2, Alternative II. The heat source is smaller (5500 W per package), so smaller temperature drops would be expected. If the borehole wall is at 150°C, however, the overpack outer wall will be at 197°C, its inner wall will be at 200°C, and the canister surface will be at 236°C for  $\epsilon = 0.61$ , making the overall drop 116°C versus 119°C for CHLW. This small difference requires some comment. First, although the heat conducted is only 58% of that for CHLW package, the SF2 package is somewhat smaller, increasing the surface flux for the SF2 canister to 63% of that for the CHLW canister.

Second, the radiant heat transfer is extremely sensitive because of the fourth power of temperature and because the calculation involves taking differences of two large numbers of comparable magnitude. Thus, a 10°C rise in  $T_1$  (about 2%) results in a 63% increase in the contribution of the radiant energy term:

$$0.61(470.15)^4 - 0.80(423.15)^4 = 4.155\text{E}+09$$

$$0.61(480.15)^4 - 0.80(423.15)^4 = 6.773\text{E}+09, \text{ or } +63\%.$$

If the emissivity of the canister is reduced, the temperature drop is increased. A solution is not obtained for  $\epsilon = 0.21$  and a borehole temperature of 150°C, but for  $\epsilon = 0.305$ , one-half the value first assumed, the drop across the canister/overpack air gap is 100°C, and the overall drop is 150°C.

## Case 2. Heat transfer within the waste packages

### A. CHLW packages

We assume the radionuclides within the CHLW packages are distributed uniformly and hence there is a uniform heat generation rate, decaying with time. The maximum temperatures within these packages should occur along the axial centerline. Assuming the packages to be infinite cylinders would lead to conservatively high values for these temperatures since this assumption neglects axial heat conduction. At steady state, which also means assuming no decay in heat

generation, the temperature distribution within the cylindrical packages is obtained from a general solution in Carslaw and Jaeger:<sup>a</sup>

$$31. \quad T - T_s = (q_v/4k)(a^2 - r^2),$$

where

$T$  = temperature at radial distance  $r$ , K

$T_s$  = surface temperature, K

and  $a$  = radius of the cylindrical waste package, m.

<sup>a</sup>Reference 5, page 191.

The volumetric heat generation with the CHLW package is

$$32. \quad q_v = (9500\text{W})/(\pi(0.280)^2(4.09)) = 9430 \text{ W/m}^3$$

and the temperature difference between the centerline and the surface is:

$$33. \quad T - T_s = (9430)(0.280)^2/(4)(1.1 \text{ W/m-K}) = 168.0^\circ\text{C}.$$

Thus, the centerline temperature for CHLW could be  $168 + 384$ , or  $552^\circ\text{C}$ .

ONWI-438 states that the waste form centerline temperatures for Alternate II should be slightly lower than those for Alternate I, which are said to peak at  $475^\circ\text{C}$ . The analysis in this report indicates reasons to conclude that at least for the initial years of emplacement the CHLW temperatures could very well exceed  $500^\circ\text{C}$ .

#### B. SF2, Alternate II, Packages

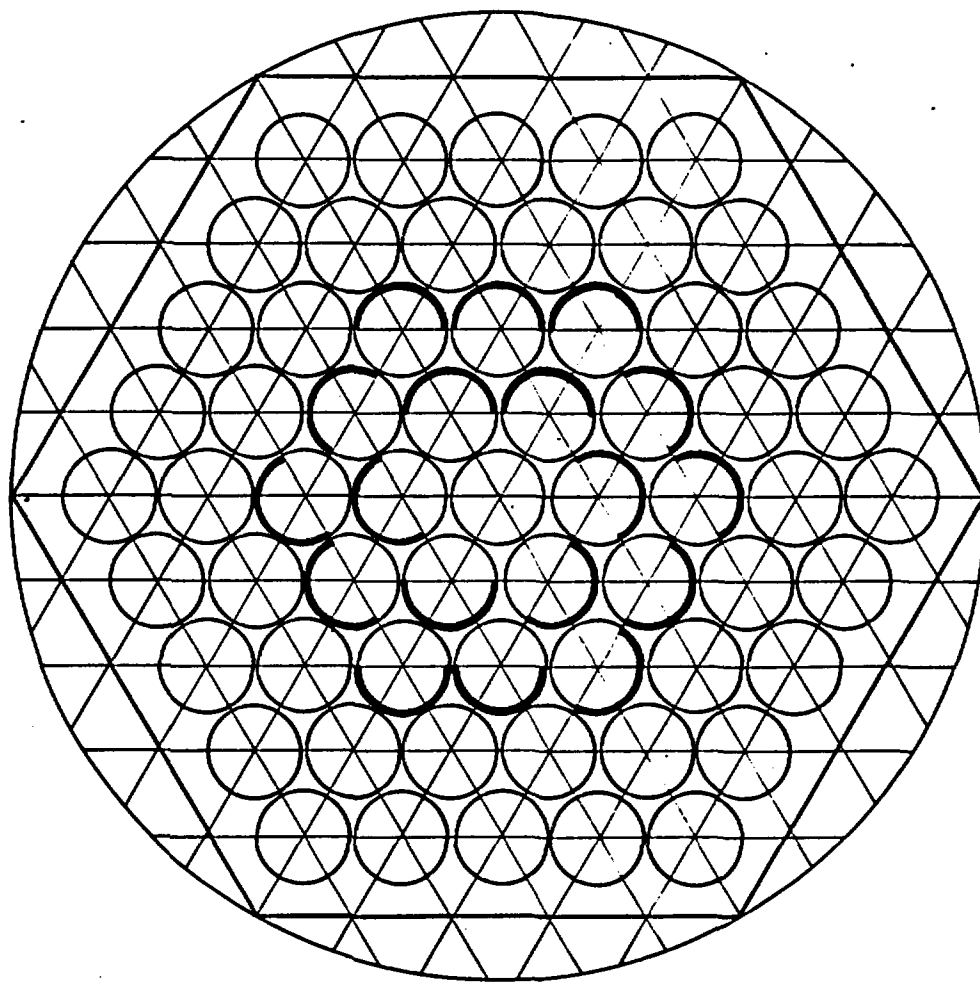
This case is more difficult to assess because it involves radiant heat transfer between close-packed fuel rods on a triangular pitch. ONWI, in fact, states<sup>b</sup> that "it is recognized that rod bundle temperature prediction is an area that requires upgrading as the package design continues." However, through simplifying assumptions an estimate can be made of the magnitude of the temperature difference between the central rod and a peripheral rod.

The first simplification is to assume a symmetric geometry as indicated in Figure 1. There is a single central rod surrounded by successive layers containing 6, 12, 18, ... rods on a close triangular packing. The total number of rods is given by:

$$\begin{aligned} 34. \quad N_r &= 1 + 6 + 12 + 18 + \dots \\ &= 1 + 6(1 + 2 + 3 + \dots) \\ &= 1 + 6n(n+1)/2, \end{aligned}$$

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<sup>b</sup>Reference 3, page 328.



**FIGURE 1**  
**LAYOUT FOR CYLINDRICAL RODS ON CLOSE HEXAGONAL PACKING**

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where  $n$  is the number of layers of rods. For  $n = 29$ , the total number in this arrangement would be 2611, which is close to the 2640 rods contained in ten PWR assemblies.

The next simplification is to assume that the temperature in any transverse cross section of any rod is constant. We also assume that all heat is transferred only radially and only by radiation. The last two terms of equation 26 then drop out, leaving:

$$35. \quad Q/A = F\sigma (\epsilon T_1^4 - \alpha T_2^4)$$

Values for  $\alpha$  are often estimated from the values of emissivity at the temperature  $T_2$ . Since we do not expect large temperature differences between adjacent layers of rods, we may further simplify to:

$$36. \quad Q/A = F\sigma\epsilon (T_n^4 - T_{n+1}^4).$$

As a compensation, we might evaluate  $\epsilon$  at the average of  $T_n$  and  $T_{n+1}$ . We do, however, expect a significant temperature difference between the central rod and the peripheral rods. As a first approximation, we assume  $T_1$ , the temperature of the central rod, is 500°C and choose an appropriate value for  $\epsilon$  at this temperature.

The fuel rods are made of Zircaloy, whose surface is probably at least partially oxidized. Highly polished metals have low emissivities, like 0.018 for copper, which would mean high temperatures to obtain the desired heat transfer. Oxidized metals, like iron, as noted above in Case 1.A.1 may have emissivities in the range 0.61 to 0.85. Because of the uncertainty of  $\epsilon$ , we shall later examine the sensitivity of calculated temperatures to the value of  $\epsilon$ . To start,  $\epsilon$  will be set at 0.5.

On the basis of the previous examination of radiant heat transfer across the overpack/borehole gap, we will also set the view factor  $F$  at 1.0. Solving for the temperature difference to transfer the heat generated by the central rod to the surrounding six, we have:

$$\begin{aligned}
37. \quad T_0^4 - T_1^4 &= Q/A\sigma\epsilon \\
&= \frac{(2.083 \text{ W/rod})}{\pi(0.95)(365.8)} \times \frac{1 \text{ cm}^2 - \text{K}^4}{0.50(5.67\text{E}-12)\text{W}} \\
&= (6.730\text{E}+08) \\
&= B, \text{ a constant}
\end{aligned}$$

For this we find  $T_1$  is 499.6°C.

As we examine successive layers, we find we do not need to calculate all the intermediate temperatures. For the second layer, the heat to be transferred is that from seven elements and the radiating surface available is the outer portion of the outer six rods. From Figure 1, this area is identified as half the surface of six rods plus six 60° sectors, or the equivalent of the cylindrical surfaces of four rods. Applying equation 34,

$$38. \quad T_1^4 - T_2^4 = \frac{7}{4}B.$$

The third layer yields

$$39. \quad T_2^4 - T_3^4 = \frac{19}{7}B$$

The fourth layer yields

$$40. \quad T_3^4 - T_4^4 = \frac{37}{10}B$$

We see that the number of tubes generating heat is that given by equation 31 and the number of equivalent rod cylindrical areas is  $(1 + 3n)$ . Summing over  $n$  layers yields:

$$41. \quad T_0^4 - T_{n+1}^4 = \sum_0^{29} \frac{1+3n(n+1)}{1+3n} B$$

In this summation, setting  $n = 0$  yields the temperature difference between the central rod and the inner surface of the first layer;  $n = 1$ , between the outer surfaces of the first layer and the inner surfaces of the second layer; and so on. With 29 layers, the summation must be carried to  $n = 28$ . A different calculation must be made for the temperature difference between the outer surfaces of layer 29 and the inner surface of the canister because (1) the geometry, and hence the view factor, is different and (2) a different emissivity should be considered for the canister metal. The summation is then:

$$\begin{aligned}
 42. \quad T_0^4 - T_{29}^4 &= B(1 + \frac{7}{4} + \frac{19}{7} + \frac{37}{10} + \dots + \frac{2437}{85}) \\
 &= 426.05B \\
 &= 2.867E+11
 \end{aligned}$$

For  $T_{29} = 100^\circ\text{C}$ ,  $T_0 = 470.68^\circ\text{C}$

The summation can be approximated to within 2% for  $n$  greater than five by:

$$\begin{aligned}
 43. \quad T_0^4 - T_{n+1}^4 &= B \sum_0^n [n + \frac{2}{3} + \frac{1}{3(3n+1)}] \\
 &\sim B \sum_0^n [n + \frac{2}{3}] \\
 &= B \frac{(n+1)}{2} (n + \frac{4}{3})
 \end{aligned}$$

Thus, if the canister is at  $100^\circ\text{C}$ , the central rod could be at  $470^\circ\text{C}$ . This analysis does not, however, reveal how soon such temperatures would be reached.

We should also check the effect of emissivity. Table 8 shows relatively little reduction in  $T_0$  (about  $100^\circ\text{C}$ ) if the emissivity could be increased to 0.9, but a sharply increasing  $T_0$  as the emissivity is reduced.

The foregoing calculation shows that cylindrical packing can lead to excessively high temperatures at the axis of the packing. DOE has recognized this problem and has proposed packing designs which (a) eliminate some of the rods at and near the axis of the bundle and (b) include radial webs to help conduct heat from the center of the bundle<sup>a</sup>. These measures may not, however, be sufficient. What our analysis suggests is that the number of layers of rods must be limited to keep the central temperature below criteria such as those in Table 2.

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<sup>a</sup>Reference 3, Section 6.4



Table 8  
Sensitivity of Central Rod Temperature to Emissivity

<u>Emissivity</u>	<u>T<sub>0</sub>, Central Rod Temperature, °C</u>
1.0	362.0
0.9	377.0
0.7	415.0
0.5	470.7
0.3	566.6
0.2	653.2
0.1	824.8

Basis: Temperature of peripheral rods is 100°C

#### Host Rock Thermal Concerns

##### 1. Brine migration

We may next proceed to a brief discussion of the impact of the foregoing thermal findings on selected concerns regarding the host rock. We first consider the water content of salt deposits.

It is known that salt deposits generally contain small amounts of water, typically less than 5 volume percent. This water may be present as pockets of brine or as brine inclusions that may also contain a gas phase. It has been shown experimentally, Reference 10, that all-liquid brine inclusions can migrate up a thermal gradient while liquid/gas inclusions migrate down a gradient. The mechanism for the former is thought to be increased solution on the hotter end of the inclusion and decreased solution (precipitation) on the colder end. This mechanism is somewhat supported by evidence that larger inclusions migrate faster, which might indicate that below a certain size the inclusions cannot sustain an internal temperature or concentration gradient. In the case of a brine/gas inclusion, it is argued that water evaporates into the gas space at the hotter end of the inclusion and condenses out at the

colder end. This results in crystallization of salt at the hot end and dissolution at the colder more dilute end. This may be a net effect since presumably the opposite transfer is occurring in the brine phase. The migration of all-liquid inclusions appears dependent on the enhancement of the solubility of salt at surface defects in the crystal structure. The velocity of intracrystalline migration is correlated by the Jenks-Claiborne equation, Reference 11:

$$41. \quad \log (V/G) = 0.00656 T - 0.6036,$$

where  $V$  = brine migration velocity, cm/y

$G$  = thermal gradient, °C/cm

and  $T$  = temperature, °C

The velocity appears sensitive to both temperature and temperature gradient, as shown in Table 9.

The velocity is, of course, proportional to the gradient, but increasing the temperature by 200°C, which would be a factor of only 1.54 on the Kelvin scale, increases the velocity at a given gradient by a factor of 20.

For an array of identical packages, it is necessary to consider migration in three directions. Halfway between adjacent packages within a tunnel, the temperature gradients due to each package are equal but opposite. A particular inclusion located off this center line would be subject to a net gradient, so it would migrate toward the nearer package. A similar consideration applies to corresponding packages in adjacent tunnels, although the gradients would be smaller. There is also a gradient in the Z or vertical direction.

As shown previously in this analysis, it is likely the Z-direction will gradually emerge as the main direction for thermal conduction. We believe it is similarly likely that any brine migration will be mainly in the Z-direction. For CHLW packages, the amount of brine available per package is estimated as  $0.05 (10)(31.6)$  or  $15.8 \text{ m}^3/\text{m}$  of height, assuming 5 volume % brine inclusions. If we consider the temperature pulse due to the waste packages penetrates  $\pm 10 \text{ m}$  in the Z-direction, then each CHLW package would have available  $(24.57)(15.8)$  or  $388 \text{ m}^3$  of brine.

Table 9  
Sensitivity of Brine Migration Velocity, cm/y

<u>Gradient, °C/cm</u>	<u>Temperature, °C</u>	
	<u>100</u>	<u>300</u>
1	1.1	23
10	11.3	231

However, temperatures above 100°C are likely, so before the repository is backfilled (when the ambient pressure is essentially atmospheric), any water in the salt near the waste packages will probably be vaporized. Some thought needs to be given, therefore, to the manner in which this water vapor escapes. If the permeability to flow is low, pressure could build up in the inclusions. Some structures might have sufficient tensile strength to contain these pressures, but others, since lithostatic pressures do not prevail in the early years of operation, might disintegrate into rubble. Especially vulnerable would be borehole and tunnel walls. Depending on the amount of inclusions present, clouds of steam might emerge into the passageways. The question of whether this steam would contain any induced radioactivity should be investigated. As the temperature of the salt continues to rise, the interface between the advancing heat pulse and the migrating inclusions will move farther away from the waste packages. The interface may have some thickness and it could be a zone of rapid cycling of wetting and drying. Such behavior in a boiler tube greatly reduces tube life and is normally avoided in designing vaporizers. Here, it could disrupt crystal structure. As the radionuclides decay, heat generation is reduced and the interface described above moves back toward the packages. Whether brine can then contact the packages depends on what happened to the water and also to the permeability of the salt.

1. Did water escape as vapor or gas permanently?
2. How much water was decomposed radiolytically?
3. Were the inclusions driven so far away that they cannot return to the packages?

4. Was the effective porosity of the salt reduced?
5. Are the thermal gradients between packages still identical?
6. Will the returning brine become more concentrated or change in composition as it equilibrates with brine solids left from the brine initially vaporized?

## 2. Bentonite stability

In the case of bentonite backfill as in a basalt site, on the one hand, water is necessary to achieve maximum packing density,<sup>a</sup> which is desirable for reducing permeability and increasing thermal conductivity. On the other hand, water near the waste packages is undesirable from the aspect of corrosion. In any case, it appears that initially the backfill will probably be dried rapidly, possibly accompanied by hydrothermal alteration of the bentonite. If the water vapor cannot escape readily, the backfill will be fluffed up by the expanding vapor and may even be blown out of the boreholes.

In a survey of the literature on bentonite hydrothermal stability, Brookhaven<sup>b</sup> reported that:

1. Some alteration of montmorillonite (bentonite) occurs at temperatures as low as 40°C.
2. At 250°C, 80% of the smectite layers could be converted to illite in 167 days.

Other evidence<sup>c</sup> shows:

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<sup>a</sup>Reference 12, Figure 5.8, page 226

<sup>b</sup>Reference 12, page 202-203

<sup>c</sup>Reference 13

3. Rapid degradation in about seven days can occur at temperatures as low as 150°C for a water/clay mass ratio of 0.1.
4. Higher temperatures are required for the same effect with larger or smaller mass ratios.
5. Water vapor at temperatures above 150°C can increase permeability by five orders of magnitude.
6. The permeability is, however, not affected much on heating from ambient temperature to 150°C (Figure 4, Reference 13).
7. Loss of swelling ability and resultant increase in permeability are irreversible.

Thus, the very near field (within tens of feet) temperature and moisture profiles are extremely important to the waste package design and ultimate performance.

### 3. Corrosion rates

As a chemical reaction, the corrosion rate of a given substance should increase with temperature in a manner which is often found to be an Arrhenius relationship, of the form:

$$42. \quad -dC/dt = k_0 e^{-\Delta H/RT},$$

where  $k_0$  = the reaction rate at some reference temperature

$\Delta H$  = activation energy, calories/g-mole

$R$  = gas constant, 1.987 calories/g-mole-K

and  $T$  = absolute temperature, K.

A plot of  $\ln (dC/dt)$  vs.  $1/T$  permits evaluation of the two constants  $K_0$  and  $\Delta H$ . Table 10 illustrates the effect of temperature on reaction rate. The figures in the body of the table are values of the exponential term in equation 42.

Table 10  
Effect of Temperature on Chemical Reaction Rate

	Temperature, °C			
	0	100	200	300
$\Delta H = 20,000$	9.92-17	1.93-12	5.77-10	2.36-08
Ratio	1	19,446	299	41
Factor	-	2.68	1.77	1.45
$\Delta H = 10,000$	9.96-09	1.39-06	2.40-05	1.54-04
Ratio	1	139	17.3	6.4
Factor	-	1.64	1.33	1.15
$\Delta H = 5,000$	9.98-05	1.18-03	4.90-03	1.24-02
Ratio	1	11.8	4.2	2.5
Factor	-	1.28	1.15	1.10

Notes:

1. Figures in the body of the table opposite  $\Delta H$  values are values of  $\exp(-\Delta H/RT)$ .
2. "Ratio" means the ratio of the value of the exponential term at a given temperature to that at the next lower temperature.
3. "Factor" means the value of  $r$  in  $r^{10} = \text{"Ratio"}$ .

The problem is that corrosion can proceed by several mechanisms and is made more complex by the influence of many variables besides temperature: radiation, system composition, stress, configuration, previous history, and inhomogeneities. System composition includes both the corroding material and its environment. Data for various corrosion modes in particular cases do show Arrhenius-type temperature dependencies:

- These considerations provide ample support to the importance of determining the temperature profile in the very near field with a high degree of certainty.

Generally, the solubility of most substances in water increases with temperature. The problem of assessing the effect of temperature on solubility is complicated in the present case because radionuclides formed on alteration of a glass waste form would typically be chemically complex, hydrated, amorphous compounds (Reference 15, page 4.45). For the glass itself, this same reference, page 1.34, states that Arrhenius-type plots are obtained for glass dissolution processes

like diffusion-controlled dealcalization and matrix corrosion, with an activation energy of the order of 20,000 calories/g-mole. Solubility correlations for low flow conditions also involve an exponential term. The  $\Delta H$  here is the enthalpy of dissolution, which may have values of about 3,000 calories/g-mole.

Transport of radionuclides may also be increased by changes in liquid boundary layers parameters such as diffusion coefficients and film thicknesses due to elevation of temperature (Reference 16, page 284). Local pore velocities, according to data from Rockwell Hanford Operations for basalt, could increase from  $3.2E-03$  m/y to  $0.64$  m/y, a factor of 200, as the temperature increases from an ambient  $57^{\circ}\text{C}$  to  $180-250^{\circ}\text{C}$ . The groundwater travel time to the edge of the host rock might then be only about 30 years. The same reference, however, states that the travel time from the host rock to the Columbia River would be about 200,000 years and also that concentrations of migrating radionuclides would be attenuated by dispersion in the far field. We concur with the conclusion that uncertainties in solubilities and transport rates should be reduced, but favor clearing up uncertainties in the temperature profiles first.

## 5. Mechanical properties

The temperature profiles in the near-field are extremely important to rock properties in interest in mechanical design. Perhaps the first impact of a temperature rise is volumetric expansion of the host rock. A second effect is acceleration of the creep rate. These and other effects need to be considered in the context of  $300-500^{\circ}\text{C}$  rather than  $100-200^{\circ}\text{C}$ . It is outside the scope of this paper to evaluate these effects quantitatively, but some qualitative observations may be made. The coefficient of linear expansion of salt is  $40E-06$  per  $^{\circ}\text{C}$  (Reference 7, page 114). Basalt, tuff and various rocks have coefficients that are only 10 to 25% of this value. For salt, every  $100^{\circ}\text{C}$  rise in temperature means an expansion of 0.4 cm per meter of salt. While the initial response may be an increase in compressive load on the salt, subsequently there may be relief of these loads due to void closure by creep or differential movement with consequent fracturing and increase in permeability. The coefficient of expansion of liquid water in the range  $20$  to  $210^{\circ}\text{C}$  as estimated from the specific volume of water in steam tables appears to be an order of magnitude larger so the compressibility of water relative to rock needs to be considered. This is not going



to be easy as field tests with in situ heaters gave poor correlations between predicted and observed deformations in efforts to determine coefficients of linear expansion (Reference 7, page 114).

### Conclusions

1. Heat loadings for the Alternate II package designs appear to result in undesirably high temperatures within and near the waste packages.
2. Previous models treat the heat transfer as a problem in radial conduction whereas it appears that within a few years after emplacement axial conduction is the dominant mode.
3. The host rock serves basically as a storage medium for the total decay heat which is conducted to the Earth's surface only after a very long time.
4. Thermal properties, and especially thermal conductivities, of the materials of construction may be considered as the principal design parameters. In the case of air gaps in the package design, emissivities are also important.
5. Scenarios based on the presence of liquid water in contact with the waste package components do not appear applicable until possibly a few hundred years after emplacement.

### Recommendations

1. Make a more definitive review of temperature profiles within and near the waste package using three-dimensional models.
2. Develop scenarios for the water content of the very near field for various times after emplacement.
3. Review internal corrosion and degradation of waste canister due to elevated temperatures.

4. See that proposed test programs for determining thermal response of candidate repository media are representative of the actual repository conditions.

## REFERENCES

1. 10 CFR Part 60, Disposal of High-Level Radioactive Wastes in Geologic Repositories, January 1, 1984.
2. Draft Statutory Environmental Assessment, Lavender Canyon, Revision 5. December 1984, Department of Energy, Washington, DC.
3. Westinghouse Electric Corporation, April 1983. Engineered Waste Package Conceptual Design: Defense High-Level Waste (Form 1), Commercial High-Level Waste (Form 1), and Spent Fuel (Form 2) Disposal in Salt. ONWI-438, Battelle, Office of Nuclear Waste Isolation, Columbus, OH.
4. Wang, I.S.Y., D.C. Mangold, R.K. Spencer, C.F. Tsang. Thermal Impact of Waste Emplacement and Surface Cooling Associated with Geologic Disposal of Nuclear Waste, NUREG/CR-2910, Lawrence Berkeley Laboratory, Berkeley, CA, March 1983.
5. Carslaw, H.S., and J.C. Jaeger. Heat Conduction in Solids. Oxford University Press, 1959, 2nd edition.
6. Isherwood, D. Geoscience Data Base Handbook for Modeling a Nuclear Waste Repository, NUREG/CR-0912, Volume 2, Lawrence Livermore Laboratory, Livermore, CA, January 1981.
7. Curtis, R.H. and R.J. Wart. Parameters and Variables Appearing in Repository Design Models, NUREG/CR-3586, Acres American Incorporated, Buffalo, NY, December 1983.
8. Rohsenow, W.M. and J.P. Hartnett, editors. Handbook of Heat Transfer. McGraw-Hill Book Company, New York, NY.
9. Lange, N.A., editor. Handbook of Chemistry, 6th edition. Handbook Publishers, Inc., Sandusky, OH, 1946.

10. D.R. Olander, A.J. Machiels, and S. Yagnik. Thermal Gradient Migration of Brine Inclusions in Salt. ONWI-208, Regents of the University of California, College of Engineering, Berkeley, CA, October 1980.
11. Jenks, G.H. and H.C. Claiborne. Brine Migration in Salt and Its Implications in the Geologic Disposal of Nuclear Waste. ORNL-5818, Oak Ridge National Laboratory, Union Carbide Corporation, Oak Ridge, TN, 1981.
12. Soo, P., editor. Review of DOE Waste Packaged Program. NUREG/CR-2482, Volume 3. Brookhaven National Laboratory, Upton, NY, March 1983.
13. Couture, R.A. Rapid Increases in Permeability and Porosity of Bentonite-Sand Mixtures Due to Alteration by Water Vapor. In "Scientific Basis for Nuclear Waste Management VIII," C.M. Jantzen, J.A. Stone, R.C. Ewing, editors, Materials Research Society Symposium, Boston, MA, November 1984.
14. Soo, P., editor. Review of DOE Waste Package Program. NUREG/CR-2482, Volume 5. Brookhaven National Laboratory, Upton, NY, August 1984.
15. Mendel, J.E., compiler. Final Report of the Defense High-Level Waste Leaching Mechanisms Program. PNL-5157. Pacific Northwest Laboratory, Richland, WA, August 1984.
16. Waste Isolation Systems Panel, T.H. Pigford, Chairman. A Study of the Isolation System for Geologic Disposal of Radioactive Wastes. National Research Council DOE Contract DE-AT01-80NE93031. National Academy Press, Washington, DC, 1983.