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subject: An Evaluation of Heat Generation Processes for the WIPP

### 1.0 Introduction

Nuclear criticality, exothermic reactions, and radioactive decay are possible sources of heat in the WIPP repository. Nuclear criticality has been eliminated from performance assessment calculations on the basis of low probability (see SNL Summary Memo of Record RNT-1). This memo discusses possible heat generating processes at the WIPP and the potential magnitude of the temperature increases they might induce. Heat from exothermic reactions is discussed in Section 2 and heat from radioactive decay is discussed in Section 3.

Conclusions are provided in Section 4. In summary, soon after disposal concrete hydration in the panel closures and shaft seals will give rise to temperature increases lasting a few decades. Heat from radioactive decay will generate a maximum temperature increase of less than 2°C above the ambient temperature (about 27°C) within 100 years after disposal. A number of potential exothermic reactions other than concrete hydration have been identified. These reactions are brine limited and will cause only minor perturbations to the temperature distribution within the disposal system. The maximum calculated rate of brine inflow to a waste disposal panel, and thus, the maximum exothermic reaction rates, occur for the S2 scenario involving an E1 drilling event at 350 years after disposal. By the time such a drilling event takes place heat generation from concrete seal hydration and radioactive decay will have decreased substantially, and the temperatures in the disposal rooms will have reduced to close to initial values. Note that active institutional controls are expected to prevent drilling within the controlled area for 100 years after disposal. Thus, exothermic reactions following a drilling intrusion into a waste disposal panel will be the only potentially significant heat generating processes at the time of a drilling intrusion.

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The maximum temperature that could be achieved in the panel following a drilling intrusion at 350 years occurs as a result of aluminum corrosion; this reaction could result in a maximum temperature increase of about 6°C two years after the drilling event. This predicted value of temperature increase is based on several conservative assumptions. For example, it is assumed that no aluminum corrosion has occurred prior to the drilling event and that all the brine introduced to the waste panel is available for aluminum corrosion. In reality some aluminum corrosion is likely to have occurred prior to the drilling event, reducing the volume of aluminum available for the reaction, and other reactions with lower reaction enthalpies (such as backfill hydration) or lower reaction rates (such as microbial degradation) will compete with aluminum corrosion to consume brine resulting in a smaller temperature increase. Based on similar conservative assumptions, backfill hydration could result in a maximum temperature increase of less than 5°C. These maximum heat generation rates resulting from aluminum corrosion and backfill hydration could not occur simultaneously because they are limited by brine availability. Thus, the temperature rise of 6°C represents the maximum that could occur as a result of any combination of exothermic reactions occurring simultaneously.

## 2.0 Heat From Exothermic Reactions

Exothermic reactions in the repository will liberate heat resulting in elevated temperatures. The magnitude and duration of this temperature increase will depend on the amount and rate of energy release, the geometry of the heat source, the thermal conductivities of the surrounding rocks, and any influence of groundwater or brine flow on heat transport.

In the WIPP a range of different types of reactions will occur, including corrosion, microbial degradation, waste dissolution, and concrete and backfill hydration, and these will liberate different amounts of heat at different times. The amount of heat liberated by the different reactions will depend on the extent of reaction that occurs (for example, how much gas generation or concrete hydration takes place), and the enthalpy of the reactions. The former will depend on the inventory of materials emplaced in the WIPP, and the subsequent chemical evolution of the repository system. The latter can be assessed by considering typical enthalpies for the reaction types of interest.

Enthalpies of reaction,  $\Delta_r H^\circ$ , for example reactions representing processes that may take place in the repository are given in Table 1 (from SNL Summary Memo of Record GG-7, SP-7). The reactions shown are based on the chemical conditions expected in the WIPP. Note that negative values for  $\Delta_r H^\circ$  indicate that the reactions liberate heat as they progress from left to right.

Table 1. Enthalpies of reaction,  $\Delta_r H^\circ$ , for the WIPP

Reaction	Example Stoichiometry and Standard Enthalpies of Formation, $\Delta_f H^\circ$ (kJ/mol)	Reaction Enthalpy $\Delta_r H^\circ$ (kJ/mol)	Data Source
Backfill hydration	$\Delta_r H^\circ$ $\text{MgO}_{(s)} + \text{H}_2\text{O}_{(l)} = \text{Mg}(\text{OH})_{2(s)}$ -601.7    -285.8    -925.5	-38	Krauskopf, 1982, 561
Backfill carbonation	$\Delta_r H^\circ$ $\text{Mg}(\text{OH})_2 + \text{CO}_2(g) = \text{MgCO}_3 + \text{H}_2\text{O}$ -925.4    -393.1    -1110.3    -285.8	-77	Drever, 1982, 351-256
Microbial degradation	$\Delta_r H^\circ$ $\text{C}_6\text{H}_{10}\text{O}_5 + \text{H}_2\text{O} = 3\text{CH}_4 + 3\text{CO}_2$ -805.5    -285.8    3 x (-74.4)    3x (-393.5)	-312	Lide, 1994, 5-16 to 5-37
Aluminum Corrosion	$\Delta_r H^\circ$ $\text{Al} + 3\text{H}_2\text{O} = \text{Al}(\text{OH})_3 + 1.5\text{H}_2$ 0    3x(-285.8)    1291.9    0	-434	Drever, 1982, 351-256
Anoxic corrosion of steel	$\Delta_r H^\circ$ $\text{Fe}_{(s)} + 2\text{H}_2\text{O}_{(l)} = \text{Fe}(\text{OH})_{2(s)} + \text{H}_{2(g)}$ 0    2 x (-285.8)    -569.0    0	+2.7	Wagman et al., 1982, Table 41
Waste dissolution	$\Delta_r H^\circ$ $\text{UO}_{2(s,cr)} + 2\text{H}_2\text{O}_{(l)} = \text{U}(\text{OH})_{4(aq)}$ -1085.0    2 x (-285.8)    -1655.8	+0.9	Grenthe et al., 1992, Table III-1
Concrete hydration	$\Delta_r H^\circ$ $\text{CaO}_{(s)} + \text{H}_2\text{O}_{(l)} = \text{Ca}(\text{OH})_{2(s)}$ -635.1    -285.8    -986.1	-65.2	Wagman et al., 1982, 2-26

Even though there is uncertainty surrounding the extent of reactions that will occur, the reaction enthalpies indicate that the thermal effects of anoxic corrosion of steel and the waste dissolution reaction will be endothermic and will be of low consequence to the performance of the disposal system. However, the other reactions shown in Table 1 have the potential to evolve significant amounts of heat. The potential effects of these processes on the temperature within the disposal system are discussed below.

## 2.1 Backfill Hydration

Potential temperature increases in the repository as a result of exothermic backfill hydration reactions have been evaluated by Wang (1996, attachment to this memo). In his analysis, Wang (1996) made the following assumptions:

- The reaction will proceed rapidly so that the rate of heat generation will be

controlled by brine availability.

- All brine entering a waste disposal panel will contact and react with the backfill in the panel uniformly.
- All of the emplaced backfill will undergo hydration.

The maximum calculated rate of brine inflow into a panel (about 200 m<sup>3</sup>/year) occurs for the S2 scenario involving an E1 drilling event at 350 years after disposal. The molar density of water is 5.56x10<sup>4</sup> moles/m<sup>3</sup>, and thus, the reaction rate of backfill hydration in the panel will be 1.1x10<sup>7</sup> moles/year. Based on the reaction enthalpy shown in Table 1, backfill hydration will generate a thermal load of about 13 kW. There will be about 2x10<sup>8</sup> moles MgO emplaced per panel and thus the reaction could continue for about 20 years if sufficient brine was available.

Wang (1996) estimated the maximum temperature that could be generated by backfill hydration within a panel. Assuming heat loss will occur by conduction through the salt forming the roof and floor of the panel and that heat losses through the side walls are negligible, Wang (1996) calculated that the maximum temperature rise in a panel, as a consequence of backfill hydration following a borehole intrusion and subsequent brine inflow, would be about 4.5°C.

## 2.2 Backfill Carbonation

Wang (1996) also estimated the potential temperature rise that could occur as a result of backfill carbonation. Wang (1996) assumed that the reaction will be limited by microbial CO<sub>2</sub> production; the maximum rate of CO<sub>2</sub> production is 2.9x10<sup>5</sup> moles/year. Based on the reaction enthalpy shown in Table 1, backfill carbonation will generate a thermal load of about 0.7 kW. About 3.6x10<sup>7</sup> moles CO<sub>2</sub> could be produced in a single panel (see Wang, 1996) and thus the reaction could continue for about 125 years. Wang (1996) estimated the maximum temperature that could be generated by backfill carbonation within a panel to be about 0.6°C.

## 2.3 Microbial Degradation

Wang (1996) estimated the maximum reaction rate for microbial degradation in a panel to be about 1x10<sup>5</sup> moles/year and the inventory to be about 1.2x10<sup>7</sup> moles C<sub>6</sub>H<sub>10</sub>O<sub>5</sub> per panel. Thus, the reaction could continue for about 120 years. Based on the reaction enthalpy shown in Table 1, microbial degradation will generate a thermal load of about 1 kW. Wang (1996) estimated the maximum temperature that could be generated by microbial degradation within a panel to be about 0.8°C.

## 2.4 Aluminum Corrosion

Wang (1996) determined that the rate of corrosion of aluminum will be controlled by brine availability. From Table 1, the reaction rate of aluminum corrosion in the panel will be about  $0.4 \times 10^7$  moles/year, assuming a brine inflow rate of  $200 \text{ m}^3/\text{year}$  ( $1.1 \times 10^7$  moles/year). About  $8 \times 10^6$  moles of aluminum will be emplaced in each panel and thus aluminum corrosion could continue for 2 years. Based on the reaction enthalpy shown in Table 1, aluminum corrosion will generate a thermal load of about 51 kW. Wang (1996) estimated the maximum temperature that could be generated by aluminum corrosion within a panel to be about  $6^\circ\text{C}$ .

## 2.5 Concrete Hydration

Concrete hydration reactions will occur in the seals and panel closures and in the waste.

### 2.5.1 Seals and panel closures

Concrete hydration reactions are known to proceed for extended periods (perhaps thousands of years). However, the rates of these reactions decrease with time and, within the WIPP, the greatest evolution of heat will occur during the short periods following emplacement of panel closures during the operational phase and following shaft seal emplacement and repository closure. A quantitative analysis of the thermal effects of emplacing large concrete seals in salt at the WIPP was made by Loken (1994), Loken and Chen (1995). Their analysis showed that the energy released by the hydration of the seal concrete could raise the temperature of the concrete to approximately  $53^\circ\text{C}$  and that of the surrounding salt to approximately  $38^\circ\text{C}$  one week after seal emplacement.

### 2.5.2 Waste

WIPP waste contains cement which is used to solidify liquids, particulates and sludges. Storz (1996) estimated that all the waste to be emplaced at the WIPP will contain a total of about  $8.5 \times 10^6$  kg of cement. This is equivalent to about  $1.5 \times 10^7$  moles of calcium oxide (CaO) per waste disposal panel, representing the cement as CaO. Although a substantial amount of hydration may occur prior to waste disposal, this process will continue at a slower rate after disposal. Disregarding the hydration that will occur prior to disposal and assuming a brine inflow rate of  $200 \text{ m}^3/\text{year}$ , the reaction rate of concrete hydration in the panel will be about  $1.1 \times 10^7$  moles/year, and the reaction could continue for about 1.4 years. Based on the reaction enthalpy shown in Table 1, concrete hydration will generate a thermal load of about 23 kW. Thus, using analyses similar to that used by Wang (1996) the maximum temperature that could be generated by concrete hydration of the waste within a panel is about  $2^\circ\text{C}$ .

### 3.0 Heat From Radioactive Decay

Radioactive decay of the contact handled CH and remote handled RH TRU waste emplaced in the repository will generate heat. The importance of heat from radioactive decay depends on the effects that the induced temperature changes would have on mechanics, fluid flow, and geochemical processes. For example, temperature increases could result in thermally induced fracturing, regional uplift, or thermally driven flow of gas and brine in the vicinity of the repository.

According to the Waste Acceptance Criteria (WAC), the design basis for the WIPP requires that the thermal loading does not exceed 10 kilowatts per acre. The WAC also require that the thermal power generated by waste in an RH TRU container shall not exceed 300 watts, but the WAC do not limit the thermal power of CH TRU waste containers.

A numerical study to calculate induced temperature distributions and regional uplift is reported in DOE (1980, pp.9-149-9-150). This study involved estimation of the thermal power of CH TRU waste containers. The DOE (1980) analysis assumed:

- All CH TRU waste drums and boxes contain the maximum permissible quantity of plutonium. According to the WAC, the fissionable radionuclide content for CH TRU waste containers shall be no greater than 200 grams per 0.21 cubic meter drum and 350 grams per 1.8 cubic meter standard waste box (in Pu-239 fissile gram equivalents).
- The plutonium in CH TRU waste containers is weapons grade material producing heat at 0.0024 watts per gram. Thus, the thermal power of a drum is approximately 0.5 watts and that of a box is approximately 0.8 watts.
- Approximately  $3.7 \times 10^5$  cubic meters of CH TRU waste are distributed within a repository enclosing an area of  $7.3 \times 10^5$  square meters. This is a conservative assumption in terms of quantity and density of waste within the repository, because the maximum capacity of the WIPP is  $1.756 \times 10^5$  cubic meters for all waste (as specified by the Land Withdrawal Act [LWA]) to be placed in an enclosed area of approximately  $5.1 \times 10^5$  square meters.
- Half of the CH TRU waste volume is placed in drums and half in boxes so that the repository will contain approximately  $9 \times 10^5$  drums and  $10^5$  boxes. Thus, a calculated thermal power of 2.8 kilowatts per acre (0.7 watts per square meter) of heat is generated by the CH TRU waste.

- Insufficient RH TRU waste is emplaced in the repository to influence the total thermal load.

Thorne and Rudeen (1980) estimated the long-term temperature response of the disposal system to waste emplacement. Calculations assumed a uniform initial power density of 2.8 kilowatts per acre (0.7 watts per square meter) which decreases over time. Thorne and Rudeen (1980) attributed this thermal load to RH TRU waste, but DOE (1980), more appropriately, attributed this thermal load to CH TRU waste based on the assumptions listed above. Thorne and Rudeen (1980) estimated the maximum rise in temperature at the center of a repository to be 1.6°C at 80 years after waste emplacement.

Sanchez and Trelue (1996) estimated the maximum thermal power of an RH TRU waste container. The Sanchez and Trelue (1996) analysis involved inverse shielding calculations to evaluate the thermal power of an RH TRU container corresponding to the maximum permissible surface dose; according to the WAC the maximum allowable surface dose equivalent for RH TRU containers is 1000 rem/hr. The following calculational steps were taken in the Sanchez and Trelue (1996) analysis:

- Calculate the absorbed dose rate for gamma-ray radiation corresponding to the maximum surface dose equivalent rate of 1000 rem/hr. Beta and alpha radiation are not included in this calculation because such particles will not penetrate the waste matrix or the container in significant quantities. Neutrons are not included in the analysis because, according to the WAC, the maximum dose rate from neutrons is 270 mrem/hr, and the corresponding neutron heating rate will be insignificant.
- Calculate the exposure rate for gamma radiation corresponding to the absorbed dose rate for gamma radiation.
- Calculate the gamma flux density at the surface of a RH TRU container corresponding to the exposure rate for gamma radiation. Assuming the gamma energy is 1.0 MeV the maximum allowable gamma flux density at the surface of a RH TRU container is about  $5.8 \times 10^8$  gamma rays per square centimeter per second.
- Determine the distributed gamma source strength, or gamma activity, in an RH container from the surface gamma flux density. The source is assumed to be shielded such that the gamma flux is attenuated by the container and by absorbing material in the container. The level of shielding depends on the matrix density. Scattering of the gamma flux, with loss of energy, is also accounted for in this calculation through inclusion of a gamma buildup factor.

The distributed gamma source strength is determined assuming a uniform source in a right cylindrical container. The maximum total gamma source (gamma curies) is then calculated for a RH TRU container containing 0.89 cubic meters of waste. For the waste of greatest expected density (about 6,000 kilograms per cubic meter) the gamma source is about  $2 \times 10^4$  curies per cubic meter.

- Calculate the total curie load of a RH TRU container (including alpha and beta radiation) from the gamma load. The ratio of the total curie load to the gamma curie load was estimated through examination of the radionuclide inventory presented in the WIPP Baseline Inventory Report (BIR) (DOE, 1995). The gamma curie load and the total curie load for each radionuclide listed in the WIPP BIR were summed. Based on these summed loads the ratio of total curie load to gamma curie load of RH TRU waste was calculated to be 1.01.
- Calculate the thermal load of a RH TRU container from the total curie load. The ratio of thermal load to curie load was estimated through examination of the radionuclide inventory presented in the WIPP BIR (DOE, 1995). The thermal load and the total curie load for each radionuclide listed in the WIPP BIR were summed. Based on these summed loads the ratio of thermal load to curie load of RH TRU waste was calculated to be about 0.0037 watts/curie. For a gamma source of  $2 \times 10^4$  curies per cubic meter the maximum permissible thermal load of a RH TRU container is about 70 watts per cubic meter. Thus, the maximum thermal load of a RH TRU container is about 60 watts, and the WAC upper limit of 300 watts will not be achieved.

Note that Sanchez and Trellue (1996) calculated the average thermal load for a RH TRU container to be less than 1 watt. Also, the total RH TRU heat load is less than 10% of the total heat load in the WIPP. Thus, the total thermal load of the RH TRU waste will not significantly affect the average rise in temperature in the repository resulting from decay of CH TRU waste.

Temperature increases will be greater at locations where the thermal power of a RH TRU container is 60 watts, if any such containers are emplaced. Sanchez and Trellue (1996) estimated the temperature increase at the surface of a 60 watt RH TRU waste container. Their analysis involved solution of a steady-state thermal conduction problem with a constant heat source term of 70 watts per cubic meter. These conditions represent conservative assumptions because the thermal load will decrease with time as the radioactive waste decays. The temperature increase at the surface of the container was calculated to be about 3°C.

In summary, analysis has shown that the average temperature increase in the WIPP repository due to radioactive decay of the emplaced CH and RH TRU waste will be about 1.6°C, with a maximum rise occurring at the center of the repository at about 80 years after waste emplacement. Temperature increases of about 3°C may occur in the vicinity of RH TRU containers with the highest allowable thermal load of about 60 watts (based on the maximum allowable surface dose equivalent for RH TRU containers).

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#### 4.0 Conclusions

Heat from exothermic reactions and radioactive decay will result in minor temperature increases in the repository. The potential temperature increases caused by these processes are summarized in Table 2.

Table 2. Maximum temperature increases at the WIPP.

Heat Source	Maximum temperature increase	Time of maximum temperature increase
Backfill hydration	4.5°C	20 years after a drilling intrusion
Backfill carbonation	0.6°C	125 years after a drilling intrusion
Microbial degradation	0.8°C	120 years after a drilling intrusion
Aluminum corrosion	6.0°C	2 years after a drilling intrusion
Concrete hydration (seals)	25°C in the shaft seal 10°C in the surrounding salt	1 week after emplacement
Concrete hydration (waste)	2.0°C	1.4 years after a drilling intrusion
Radioactive decay of CH TRU waste	1.6°C	80 years after disposal
Radioactive decay of RH TRU waste	3°C near a few containers	Within 100 years after disposal

During the operational phase and soon after disposal concrete hydration in the panel closures and shaft seals will give rise to temperature increases lasting a few decades. Heat from radioactive decay will generate a maximum temperature increase of less than 2°C within 100 years after disposal. A number of potential exothermic reactions have been identified. These reactions are brine limited and will cause only minor perturbations to the temperature distribution within the disposal system. The maximum calculated rate of brine inflow to a waste disposal panel occurs for the S2 scenario involving an E1 drilling event at 350 years after disposal. The maximum temperature that could be achieved in the panel

occurs as a result of aluminum corrosion; this reaction could result in a maximum temperature increase of about 6°C two years after the drilling event. Similarly, rapid backfill hydration could result in a maximum temperature increase of less than 5°C. These predicted values of temperature increase are based on a number of conservative assumptions. For example, the calculated temperature resulting from aluminum corrosion is based on the assumption that no corrosion has occurred prior to the drilling event and that all the brine introduced to the waste panel is available for aluminum corrosion. In reality some aluminum corrosion is likely to have occurred prior to the drilling event, reducing the volume of aluminum available for the reaction, and other reactions with lower reaction enthalpies or lower reaction rates will consume brine resulting in a smaller temperature increase.

Temperature increases of the magnitude and duration shown in Table 2 will have no significant effects on the performance of the disposal system. The effects of such temperature increases on the performance of the disposal system have been discussed in the SNL Summary Memos of Record RNT-24 (thermochemical effects), S-10 and GG-4 (thermal convection), S-11, SP-6, RM-1 (thermally-induced stress).

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Attachment: memorandum of August 19, 1996, by Yifeng Wang, to Distribution. "Evaluation of the Thermal Effect of Exothermic Chemical Reactions for WIPP Performance Assessment: A revised version," Sandia National Laboratories.

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