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Gerald Thiers

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4. ADDRESS

1261 Town Center Drive, Las Vegas, NV 89134, MS 423

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Document prepared under NLP-3-27 (Engineering Calculations) describing analyses of seepage into an emplacement drift with and without treatment of a zone of rock above the drift by injection of calcite. Calculation Title: "Evaluation of Near-Field Rock Treatment During Construction" (SR/LADS Design Feature 22)

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Ernest L. Hardin

RESPONSIBLE MANAGER NAME

RESPONSIBLE MANAGER SIGNATURE

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14. ADDRESS

1261 Town Center Drive, Las Vegas, NV 89134 MS 423

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Gerald Thiers

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G. R. Thiers

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EVALUATION OF NEAR FIELD ROCK TREATMENT DURING CONSTRUCTION (LADS FEATURE #22)

Eric Sonnenthal and Nicolas Spycher

1. Purpose

To evaluate the effect of near-field rock treatment by injection of reactive material (calcite) above the drift for the purpose of decreasing postclosure drift seepage.

2. Method

The method used for the calculation was a coupled reaction-transport numerical model for gas-water-rock interaction. This includes the mass conservation of heat, liquid and gas for thermohydrological calculations, of aqueous and gaseous species for advective and diffusive transport, and the kinetics of mineral-water reactions.

3. Assumptions

There are many assumptions that generally underlie numerical modeling of heat, fluid, and gas flow in unsaturated porous fractured media. In addition to these assumptions there are numerous assumptions underlying the calculation of mineral-water reactions, the transport of aqueous and gaseous species, and the conceptual models that are used to describe the chemical and physical systems.

The model assumptions are as follows (used throughout):

- (a) Dual permeability -- separate, but interacting fracture and matrix continua. The fracture continuum is considered as separate but interacting with the matrix continuum, in terms of the flow of heat, water, and vapor through advection and conduction. Aqueous and gaseous species are transported via advection and molecular diffusion between fractures and matrix. Each continuum has its own well-defined initial physical and chemical properties.
- (b) Due to symmetry, the system can be represented as a two dimensional zone given by the half-width of a drift to the center of the rock mass between drifts.
- (c) The drift wall is considered as a no-flux boundary, for the following reasons: First, the evolution of the chemical composition of the gas in the drift itself is unknown, and as an uncertain boundary condition would complicate any results for the calcite system. Second, for a homogeneous fracture system with constant surface infiltration, the wall would act as a capillary barrier, and percolation into the drift would not be adequately described by the model as chosen.

(d) Given assumption (c) above, the change in percolation flux into the drift by the addition of a reactive material to the region above the drift wall is described by a comparison between percolation fluxes through fractures to the drift wall for the cases of no injected material and injected calcite.

(e) Infiltration is temporally and spatially uniform.

(f) Rock thermohydrologic properties are initially spatially uniform.

(g) The thermal conductivity and heat capacity of the calcite is assumed to be the same as the host rock.

(h) The rock other than the calcite is considered unreactive. Although the reaction rates of the silicate minerals are typically much smaller than that of calcite, the long term behavior of the system would likely be affected by interaction of pore fluids with the wall rock. Without extensive sensitivity tests of differing primary and secondary mineral assemblages, and with calibration to laboratory and thermal test geochemical data, the full system response would be difficult to constrain.

(i) The chemical composition of infiltrating water and of the gas at the top boundary are assumed constant over time.

(j) Physical properties of the gas are unaffected by the partial pressure of CO_2 .

4. Use of Computer Software

The TOUGHREACT code V2.0 was used for all simulations. This version is a modified version of TOUGHREACT V1.0 (SCMS Reports, DF6 R00) that was qualified in accordance with YMP-LBNL-QIP-SI.0, Computer Software Qualification, and SI-1, Software Configuration Management, which meet the requirements of the QARD. TOUGHREACT V2.0 is undergoing qualification and will be qualified shortly. Until that time, all results must be considered "to be verified" (TBV). Spreadsheets and calculators were used for other simple calculations.

Work is documented in scientific notebooks YMP-LBNL-ELS-1 (pp. 1-21) and YMP-LBNL-YWT-NS-1 (pp. 150-154).

Data sources include the EQ3/6 V.7.2.B database (Wolery 1992[†]), which consists mostly of data from SUPCRT92 V1.0 (Johnson et al. 1992[†]) and are non-Q.

5. Calculation

[†] refers to database or model, in general

5.1 Permeability and Capillary Pressure Modification

The change in permeability with porosity was derived to be as follows:

$$k(t) = k(\text{initial}) * (\phi(t)^3 / \phi(\text{initial})^3)$$

where k is the permeability, t is the present time step, and ϕ is the porosity. This is based on the cubic law for plane parallel fractures (Sonnenthal et al. 1997, p. 7-14) to be consistent with the derivation of fracture permeabilities and unsaturated flow parameters used in the model. This is a conservative relation, as it does not consider the plugging of apertures narrower than the mean aperture. It considers only the uniform distribution of mineral precipitates or dissolution along fracture walls. The change in the matrix permeability was also assumed to follow this relation, as a conservative estimate.

Changes in capillary pressure with changing permeability were accomplished through incorporation of Leverett's rule, based on the formulation described in Finsterle (1997, p. 225).

5.2 Starting Water and Gas Compositions

The initial composition of water in the rock matrix and fractures was assumed to be the same as the composition estimated by Sonnenthal et al. (1998, p. 63) for pore waters representative of the Tptpmn Unit (Table 5.2.1).

Table 5.2.1. Initial matrix and fracture water composition (Sonnenthal et al. 1998, p. 63). (DTN:LB981120001233.001).

Constituent	Concentration	Concentration
	(mg/L)	(mol/L)
pH	8.2	-
Cl	41	1.157E-03
HCO3	219	3.584E-03
Na	91	3.959E-03
Ca	27	6.738E-04

(Temperature 25 C)

Bicarbonate concentration represents total dissolved carbonate concentrations expressed as HCO_3^- .

The starting CO₂ gas partial pressure in fracture and matrix pore spaces was assumed equal to the CO₂ fugacity at equilibrium with the initial water at 25 °C and atmospheric pressure of 1 bar. It was computed to be 0.00132 bars (or approximately 1320 ppmV). This initial value of CO₂ results in a large spike of CO₂ during early boiling similar to that observed in the Drift Scale Test (DST) (Conrad 1998, p. 3-3).

5.3 Initial Mineral Assemblage

To investigate the effect of calcite precipitation and dissolution, compared to the base case thermohydrologic calculations, the rock formation surrounding the drift was assumed to be unreactive except for the calcite in the injected slurry. Three cases were considered -- a system with no chemical reactions, one with 5 percent calcite (by volume), and one with 20 percent. These values refer to the volume percentages of the effective fracture porosity. The wallrock was set to be unreactive. Only calcite was added in the simulation because any added water may not be stationary, and would be quickly evaporated during the initial heating phase. The mineral proportions were estimated, because the actual amount of the mineral phase injected would depend on the fracture aperture distribution.

5.4 Thermodynamic and Kinetic Data

Dissociation constants for aqueous species and the solubility product for calcite and CO₂ gas were taken from the EQ3/6 V7.2.B database (Wolery 1992[†]), which consists mostly of data from SUPCRT92 V1.0 (Johnson et al. 1992[†]). The calcite molar volume was also taken from the SUPCRT92 V1.0 database.

Calcite was assumed to precipitate and dissolve under kinetic constraints. A rate constant of 1×10^{-11} mol/m²/s and activation energy of 41.87 kJ/mol were assumed for this mineral (Glassley 1998, p. 5-70). The reactive surface area of calcite grains in the injected slurry was estimated to be 1107 cm²/g by assuming a spherical grain shape with a grain radius of 10 microns and a mass density of 2.71 g/cm³ (SUPCRT92 V1.0: Johnson et al. 1992[†]).

5.5 Numerical Mesh and Volumes of Injected Material

The numerical mesh around the heated drift showing the region of calcite injection is shown in Figure 5.5-1 (Attachment I-1). The mesh consists of 532 fracture and matrix elements. The drift diameter was set to 5.5 meters, with an interdrift spacing of 28 meters (CRWMS M&O 1998, Section 4.2.1.2, p. 4-1). The full mesh extends to the land surface and to the water table. The surface and water table elevations were taken from the FY97 unsaturated zone site-scale model (Bodvarsson et al. 1997[†]) in the potential repository area. Data used in the model can be found in the accompanying input files.

[†] refers to database or model, in general

The region of injected calcite was taken to be approximately one drift diameter above the drift wall, about 4 meters wide and extends approximately 2 meters past the drift wall. The total volume of the injection zone shown in Figure 5.5-1 (assuming a thickness of 1 meter) is approximately 22.86 m^3 , whereas the volume of the injected calcite is $3.075\text{e-}3 \text{ m}^3$ for the 5% injection and $1.23\text{e-}2 \text{ m}^3$ for the 20% injection case. This assumes a fracture porosity of 0.00263, which is based on the value calculated for the Tptpmn for the UZ site scale model (Sonnenthal et al. 1997, p. 7-20), then increased by a factor of ten to capture the effect of flow through fractures less than 1 m in length that are not considered in the site-scale model. This is justified by the even larger calculated effective porosities in the range of 0.006 to 0.018 that are based on gas tracer measurements (Freifeld and Tsang 1998, p. 2-4).

The volume of water in the slurry would depend on the particular viscosity of the slurry desired, but should be kept at a minimum to maintain a high enough viscosity to prevent loss to surrounding areas by free drainage. It is assumed then that the physical properties (i.e., viscosity and density) would be determined once the amount of calcite necessary to modify the hydrological properties is ascertained. This may require multiple injections, allowing imbibition and evaporation of water and possibly some filtration, to obtain the required solid volume fractions.

5.6 Hydrological and Thermal Properties and Boundary Conditions

The level of the potential repository in the UZ site scale model at the chosen location is 1099.5 meters, placing it in the the Topopah Spring Middle Nonlithophysal Unit (Tptpmn). Hydrological and thermal properties corresponding to the Tptpmn unit were taken from Birkholzer and Tsang (1997, p. 18, 20) that were used for thermo-hydrological modeling of the DST and for thermo-hydro-chemical modeling of the DST by Sonnenthal et al. (1998[†]).

The surface and water table temperatures were assumed constant and taken from the FY97 UZ site scale model (Bodvarsson et al. 1997[†]). The surface air pressure was assumed constant and also assigned the value from the FY97 model. The infiltration rate in this area is approximately 13.36 mm/year (Bodvarsson et al. 1997[†]). Matrix and fracture elements at the water table were assumed to be nearly fully saturated. The drift wall was assumed to be impermeable. The areal heat load was applied equally to all drift wall elements, at the loading value of 85 MTU/acre as used in TSPA-VA and UZ thermohydrology (Haukwa et al. 1998, p. 18). Heat was applied to the matrix elements only.

The top boundary P_{CO_2} was set to the equilibrated value of 0.00132 bars, as described in Section 5.2. Therefore all water and gas entering the system during the post-thermal period (and over the entire 100,000 years) is equilibrated to this value. Concentrations in infiltrating water at the surface are also set to those given in Table 5.2.1, and are assumed constant over time.

5.7 Transport Parameters

The diffusion coefficient of aqueous species was estimated to be 10^{-9} m²/sec from data in CRC (1981, p. F-53). The coupled flow-transport-reaction calculation methods assume that the diffusion coefficient is the same for all aqueous species (e.g. Steefel and Lasaga 1994, p. 537). The diffusion coefficient of CO₂ in the vapor phase was estimated to be 2×10^{-5} m²/sec from data at 0°C in CRC (1981, p. F-53) extrapolated to an average temperature of 50°C using methods in Lyman et al. (1990, Equations 17-11 and 17-16, and references therein). The tortuosity was assumed to be 0.2 in fractures and matrix. This parameter cannot be exactly determined, and a value of 0.2 is within a typical range of values (e.g. Bear 1972, p. 111).

5.8 Simulations

Three simulations are described. They are: (1) a base case thermohydrological simulation with no chemical reactions. (2) A simulation with 5% injected calcite. (3) A simulation with 20% injected calcite. The simulations were run for 100,000 years. They included the transport of aqueous species given in Table 5.2.1 and of CO₂ in the gas phase, and the reaction of calcite with the aqueous phase.

6. Results

All results are TBV, as discussed in Section 4.

Results are shown for several times after the initiation of heating up to 100,000 years, and also for profiles through the injection zone. There are several ways in which to display the changing percolation flux at the drift wall. Two methods were chosen in this report, although other ways of analyzing the model output could also be made. The vertical flux entering the crown element directly at the drift wall is the first method. The second method sums all of the fluxes entering and leaving the drift wall elements. Because of the much higher permeability of the fracture continuum, any seepage would be more likely through fractures, and therefore percolation fluxes are shown for the fracture medium.

First, a comparison is made between the vertical fracture flux entering the wall element at the crown of the drift for the case of no chemical reactions and for the case of 20% calcite injected. Figure 6-1 (Attachment I-2) shows the vertical fracture flux as a function of time from 1 year to 100,000 years. The large pulse of liquid flux at ten years is due to the expulsion of liquid from the heated matrix. By 50 years the fractures are completely dry and therefore the liquid flux is zero. Both cases show nearly the same liquid flux at the wall element. At 500 years, the fractures again begin to rewet and the flux increases. Figure 6-2 (Attachment I-3) shows the same data, with only the time period from 100 years on. Here the slightly lower fluxes for the calcite injection case can be seen. By 2000 years, the fluxes have stabilized and remain nearly constant until the end of the simulation, at 100,000 years.

The difference in fluxes for the two cases ranges from about 7 to 8% between 500 and 1000 years, and about 5% thereafter.

The net flux into all fracture wall elements is shown in Figure 6-3 (Attachment I-4). This represents the difference between the flux entering and leaving the fracture medium, which basically describes the increased saturation of fractures and excess flux into the matrix at a particular time. The pattern is similar to that seen by the vertical fracture flux in Figures 6-1 and 6-2 (Attachments I-2 and I-3). Although the initial permeability decrease in the injection zone is about 50% for the 20% injection volume of calcite, the flux decrease is not as great because of the increased capillary suction caused by calcite precipitation at the drift wall. The initial permeability decrease for the 5% added calcite was about 14%, and the results are also little different from the base case thermohydrological calculations.

Partial pressures of CO₂ (P_{CO_2}) are shown after 10 years of heating in Figure 6-4 (Attachment I-5). Regions of increased P_{CO_2} are evident as a band above the drift (and calcite injection zone) and in a narrow region at the drift wall itself. In the region below the calcite injection zone, P_{CO_2} is substantially depressed due to exsolution of CO₂ during boiling. At the drift wall, there is a large increase in P_{CO_2} due to boiling in the zone away from the drift and buildup at the drift wall. The order of magnitude increase in P_{CO_2} above the calcite injection zone is similar to that observed in the DST by Conrad (1998, p. 3-3). Over longer time periods the region of elevated P_{CO_2} above the calcite injection zone becomes broader and extends nearly to the surface as the heat pipe zone extends upward.

The calcite redistribution (difference from initial amount) at 10,000 years is shown in Figure 6-5 (Attachment I-6). The greatest amount of precipitation is directly at the drift wall, with a small amount of dissolution in the calcite injection zone, and some precipitation below and to the side of the injection zone. The calcite that is dissolved out of the injection zone is carried in solution to the side of the injection zone, where it precipitates.

Profiles of dissolved and precipitated calcite as a function of radial distance (vertical, 45 degrees, and horizontal) at 10,000 years are shown in Figures 6-6 (Attachment I-7) and 6-7 (Attachment I-8) for the cases of 5% and 20% added calcite, respectively. The amounts and profiles are quite similar, because the solubility is the same for any amount of calcite, and the reaction rates are little different. It is evident that some calcite is dissolved out of the injection zone and redeposited along the side of the drift, where there was little calcite previously. Most calcite, however, is precipitated directly at the drift wall, due to dryout, and there is less dissolution in the post-thermal period.

Regions of increased calcite led to decreased permeability and increased liquid saturation due to greater capillary suction (Figures 6-8 (Attachment I-9) and 6-9 (Attachment I-10)). This occurred directly at the drift wall and near the top of the injection zone (Figure 6-9 (Attachment I-10)). The increased saturation, though, was quite small because little calcite was remobilized and permeabilities were therefore little affected. The increased saturation in

the injection zone has the effect of increasing the relative permeability, therefore counteracting some of the effect of decreased permeability.

Overall, the injection of calcite in the volumes used decreases the flux to the drift wall by less than 10%, at a maximum. However the volumes added were conservative values, and multiple injections leading to a larger volume fraction of calcite would decrease the fluxes to the drift wall. Because of the retrograde solubility of calcite with increasing temperature, calcite is a relatively stable phase in the injection zone, once the transient pulse of CO₂ caused by boiling in the rock above the drift wall has passed. Although the solubility of calcite increases in the cooler waters that percolate during the post-thermal period, these waters are close to saturation through interaction in the ambient system and the small temperature increase along the natural geothermal gradient. Thus, the dissolution of calcite out of the injection zone in the post-thermal period is relatively minor.

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8. Attachment I. Figures

Attachment I-1 Figure 5.5-1. Numerical mesh used for simulation of near field rock treatment. Zone into which calcite was injected is shaded in red. (DTN:LB981120001233.001).

Attachment I-2 Figure 6-1. Vertical fracture liquid flux in crown element, as a function of time, for the 20% injection case and the base case thermohydrology run. (DTN:LB981120001233.001).

Attachment I-3 Figure 6-2. Vertical fracture liquid flux in crown element, as a function of time (100 years to 100,000 years), for the 20% injection case and the base case thermohydrology run. (DTN:LB981120001233.001).

Attachment I-4 Figure 6-3. Net fracture liquid flux in all wall elements, as a function of time, for the 20% injection case and the base case thermohydrology run. (DTN:LB981120001233.001).

Fracture Log P_{CO_2} at 10 Years

Attachment I-5 Figure 6-4. Log partial pressure of CO_2 around drift after ten years of heating for the 20% injection case. (DTN:LB981120001233.001).

Fracture Calcite at 10,000 Years

Attachment I-6 Figure 6-5. Calcite distribution (volume percent change) around drift after 10,000 years of heating for the 20% injection case. (DTN:LB981120001233.001).

Attachment I-7 Figure 6-6. Calcite distribution (volume percent change) as a function of radial distance from drift center after 10,000 years of heating for the 5% injection case. (DTN:LB981120001233.001).

Attachment I-8 Figure 6-7. Calcite distribution (volume percent change) as a function of radial distance from drift center after 10,000 years of heating for the 20% injection case. (DTN:LB981120001233.001).

Attachment I-9 Figure 6-8. Liquid saturation distribution as a function of radial distance from drift center after 10,000 years of heating for the 5% injection case. (DTN:LB981120001233.001).

Attachment I-10 Figure 6-9. Liquid saturation distribution as a function of radial distance from drift center after 10,000 years of heating for the 20% injection case (DTN:LB981120001233.001).