High Level Waste in Salt Repositories: Experiments and Simulations of Evaporation in the Underground – 17167

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ABSTRACT

Advances in simulation of coupled heat and mass transfer in granular salt using the Finite Element Heat and Mass Transfer Code (FEHM) is presented in this paper. The modeling focuses on reproducing the results of an in-situ subsurface evaporation experiment beginning during the summer of 2015 carried out in the underground at the Waste Isolation Pilot Plant (WIPP). Run-of-mine (granular) salt was placed in a metal pan, and sample mass, as well as ambient temperature and humidity, were measured continuously on a data logger through May 2016. Changes in moisture content of the salt are interpreted from the change in mass, and these data provide insight into evaporation and water retention characteristics from salt in the WIPP underground.

To allow simulation of the experimental data, FEHM was modified to include a variable humidity boundary condition. The new humidity boundary condition can adjust streams of inflowing air to reflect fractions of air and water at prescribed temperature and pressure conditions that in turn represent relative humidity. Attributes for the flow such as temperature, air mass fraction, and humidity can be specified for inflowing air. The in-situ experiment provided data for validation of FEHM code development to accurately compute water-vapor pressure lowering due to dissolved salt and capillary pressure for pore water stored granular WIPP salt. Using this new boundary condition capabilities, a numerical representation of the pan experiment was created. The numerical domain is a 2-dimensional square grid of 30 by 30 cm axes. Salt and air properties are assigned based on measured values. A low flow rate of air is specified across the pile of salt to represent drift ventilation airflow. Humidity and temperature of the flowing air are fixed to measured values, and the simulated pile of salt is allowed to take up or release water based only on gradients in humidity and water-vapor pressure. Mass changes of the modeled salt pan were compared to measured mass changes in the bench-scale experiment. Directionality of the modeled changes tracks well with measured values, with good correlation between peak and valley inflection points. This shows that in situations with airflow where the relative humidity boundary can apply, FEHM can produce accurate values if humidity, air flow, and initial conditions are well established. Finally, this work supports phased-field-thermal testing in salt planned for the underground at WIPP that is expected to begin in early 2017.

INTRODUCTION

As part of the Department of Energy's (DOE) Environmental Management (EM) and Nuclear Energy (NE) research programs into nuclear waste repositories, Los Alamos National Laboratory (LANL) has been involved in both experiments and simulations that seek to increase our understanding of the complex and coupled processes that occur when hot waste interacts with salt formations in the underground. Specific objectives related to disposal system performance are defined in an update to the UFD Campaign Implementation Plan [1]. For 2013-2016 the research has focused on using theory, experiments, and modeling in combination with existing underground research laboratory (URL) data to assess disposal system performance, including reduction of uncertainty associated with heat-generating nuclear waste (HGNW). HGNW is defined herein as the combination of both heat-generating defense high-level waste (DHLW) and civilian-spent nuclear fuel (SNF). Thermal, hydrological, mechanical, and chemical (THMC) coupling and related modeling, as well as development of a field testing plan are discussed in this objective.

Salt is an attractive material for the disposition of HGNW because of its self-sealing, viscoplastic, and reconsolidation properties [2]. The rate at which salt consolidates and the properties of the consolidated salt depends on the composition of the salt, including its weight percent of accessory minerals and moisture content, and the temperature under which consolidation occurs. Physicochemical processes, such as mineral hydration/dehydration, salt dissolution, and precipitation significantly influence the rate of salt structure changes.

Brine and mixed-phase migration of fluids in salt is important for understanding the self-sealing behavior of a salt repository [3]. Experimental studies have demonstrated that porosity may migrate towards a thermal source for small-scale fluid inclusions within salt crystals [4]. However, in some conditions when pore space in salt is sufficiently connected for fluid migration to occur, porosity may be expected to migrate away from a heat source [5]. As saline water evaporates, salt concentrations increase until saturation is reached; as further water is removed, salt will begin to precipitate into void spaces previously occupied by water. Consequently, areas in which evaporation is occurring tend to be self-sealing, with reductions in porosity and permeability [2]. Evaporated water, however, will condense as temperature decreases, resulting in dissolution of salt farther afield and increases in porosity and permeability [6]. The water, brine, and gas content of the salt is dynamic in the subsurface at the WIPP, and will change with the temperature and humidity conditions of the forced ventilation air in the waste storage galleries and access drifts. Properly representing saturation in our modeling efforts of HGNW repositories at WIPP is critical for predicting the THMC deformation and self-sealing behavior of the salt that create a safe long-term environment for HGNW storage.

Another behavior of vapor and liquid water in salt to consider for HGNW storage is the vapor pressure lowering for saline water. Water-vapor pressure ($\mathrm{P_{wv}}$) lowering due to dissolved salt in liquid water is an important process to accurately represent in simulations of gas and water flow in porous salt. $\mathrm{P_{wv}}$ can be thought of as the tendency of liquid water to change phases. As this value approaches ambient (i.e. atmospheric) pressure, increasing amounts of water will go into the vapor phase. As temperatures increase, P_{wv} will also increase because the air has a greater capacity to hold water vapor at warmer temperatures; the temperature-P_{wv} relationship follows an exponential curve with P_{wv} increasing more rapidly as air warms (Figure 1). In contrast, increased solute concentration has the opposite effect. The addition of salt lowers the P_{wv} of saline brine compared to equivalent pure water and therefore raises the boiling point of water.

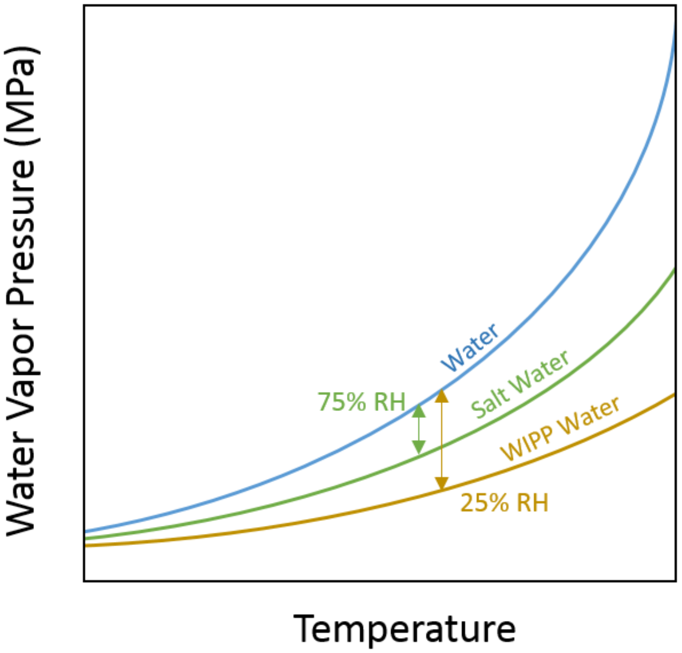

Figure 1: Conceptual model of water vapor pressure for pure water (blue), water containing salt (green), and water in granular WIPP salt (yellow).

The P_{wv} lowering tendency of water with dissolved WIPP run-of-mine (RoM) salt is greater than for salt-saturated water, and likely depends on the capillary pressure in pore spaces of RoM salt. P_{wv} for water saturated with pure salt is lowered, requiring the relative humidity of the air to be about 25 % lower than the relative humidity for a phase change to occur for pure water. With the combination of the dissolved salt (and other accessory minerals) along with the liquid-water retention caused by capillarity in granular salt, it may be expected that P_{wv} would be depressed even more than in pure salt (Figure 1). Field observation of in WIPP RoM salt suggests that as vapor pressure is lowered, the relative humidity of the air must be about 75 % lower to cause phase changes, as compared to pure water. This is a considerable decrease in the exchange between the liquid water and vapor-phase water. The added influence of salt on P_{wv} is important for retention characteristics of water within pores and the consequent dissolution and precipitation of salt in the presence of a thermal gradient.

Understanding the behaviors of these complex and coupled processes that control the self-sealing behavior of salt is important for safe design for disposal of HGNW in salt formations, so experimentation and modeling is underway to characterize these processes. Because of the complicated physics involved with the THMC changes in granular salt, numerical modeling of salt has required the addition of many capabilities to the Finite Element Heat and Mass Transfer Code (FEHM; https://fehm.lanl.gov), a hydrologic multiphase flow and transport model developed at Los Alamos National Laboratory [7, 8, 9]. Although the code was originally intended to simulate geothermal reservoirs, subsequent revisions and additions in the past 30 years have allowed it to solve for unsaturated flow, reactive chemistry, stress, and carbon dioxide. FEHM uses a finite volume method for solving the conservation of mass and momentum equations. These additions consist of a new module specifically designed for simulation of salt, as well as minor changes to the fundamental FEHM code. Subsequent code corrections have been implemented for numerical efficiency and accuracy.

To validate the new physics included for modeling salt with FEHM, we are comparing simulation results to those of simple experiments and analytical solutions to isolate specific behavior. The work presented in this paper builds on previous experimental work, code development, and associated modeling [10, 11, 12, 13, 14, 15]. This paper discusses an experiment performed at WIPP, in the URL, and shows how FEHM simulations are reproducing physical behaviors of the system. An experiment was constructed underground at WIPP in which RoM salt was placed in a metal pan, and ambient temperature, humidity, and sample mass were measured continuously from May 2015 through May 2016. Comparison of modeled to experimental results allow for validation of FEHM salt-modeling capabilities, and build confidence in results for further modeling for HGNW testing and disposal design.

DESCRIPTION

Experimental Setup

A simple, long-term experiment was constructed for observing mass fluctuations of RoM salt in response to changes in relative humidity of air in a gallery In the URL at the WIPP site. Two pans were filled with RoM salt and placed on scales to measure the mass changes for the period of July 10, 2015 to May 3, 2016. The humidity and temperature of the air in the gallery was also measured; the three measurements were recorded simultaneously every 10 minutes.

Figure 2: Salt pan experiment in WIPP salt gallery. Pans with salt piles are on left.

Model setup

Modeling of the WIPP salt pan experiment is on a 2-dimensional, 0.3 m by 0.3 m square grid (Figure 3). The grid has uniform 0.01 m spacing resulting in 960 nodes and 900 elements. Salt and air zones are defined; salt properties are as described in TABLE I. Porosity and initial saturation estimates were determined from lab measurement of RoM salt in the rock mechanics laboratory at LANL. The permeability was based on estimates of permeability of granular rocks of other mineralogies (Fetter, 2001). The salt zone is asymmetric to create the cone geometry in a radial space of the pans in the experiment as the salt was higher in the middle than the sides.

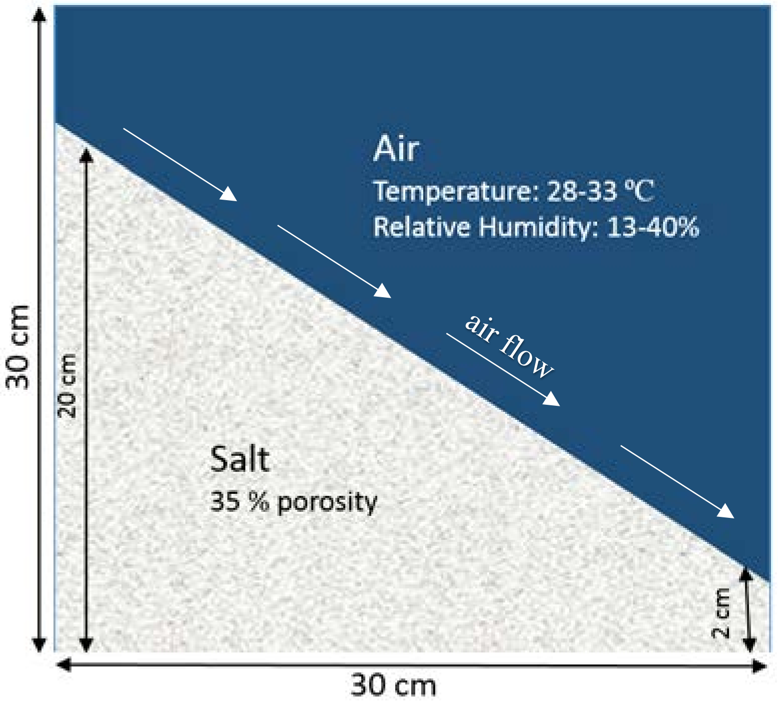

Figure 3: Model domain for salt pan experiment.

TABLE I: Salt properties used in modeling.

Property	Value	Units
Solid Density [16]	2165.0	kg/m^3
Specific Heat Capacity *	931.0	J/kg·K
Permeability ♦	1×10^{-12}	m^2
Thermal Conductivity [14]+	2.0	W/m·K
Porosity ♦	0.35	-
Initial saturation ♦	0.4	-

* Bulk value based on porosity of 35 %, $cp,bulk = (1 - \phi)cp,salt + \phi cp,air$

\+ Eq. 2-4 in *Stauffer et al.*, 2013, based on porosity of 35%

♦ Based on lab measurement

To represent the changing humidity boundary condition, the macro control statement *fxa* was implemented. This statement applies a time-variant mixing ratio of air mass to water-vapor mass. We determined the mass-fraction of air to water from the measured record of humidity in the gallery from the field measurements of temperature and humidity. For reference, a mixing ratio approaching 1 represents dry air, and as the mass of water in the vapor phase increases as the air is wetted, the ratio will decrease. The input values for *fxa* statement require simulation of flowing air. To accommodate this in the model, a pressure change of $1 \text{ x } 10^{-5}$ MPa

(10 Pa) over 30 cm was applied causing a slight inflow of air directly above the peak of the cone and a slight outflow of air directly above the low point at coordinates (30, 0.3) cm. A low flux of air with the desired water vapor mixing ratio would pass across the top of the salt cone and evaporate or condense pore water based on proportional water content of the air. The transient mixing ratio applied with *fxa*, in conjunction with measured temperature in the galley, provides FEHM the data to calculate a relative humidity boundary condition in the air.

A background run was conducted first to reach a steady-state, initial condition for subsequent simulations. A 12-day model run was conducted using laboratory-measured temperature and humidity conditions.

RESULTS AND DISCUSSION

Experiment

We observed from the RoM-salt-filled pan that increased evaporation tends to track with decreases in the relative humidity in the surrounding air. Figure 4 shows the comparison of the measured change in mass of the pan to the relative humidity of the air in the gallery. We assume that all changes in the mass of the salt are the result of evaporation and condensation of water in the liquid phase being stored in the pore spaces of RoM salt. We observed a significant drop in the mass of the pan in the first 30 days of the experiment, when the relative humidity of the air is relatively high (generally above 40 %), but we attribute this to evaporation of water from salt that was recently exposed to air. The transient salt saturation following the granular salt being newly exposed to air would explain this early-time mass loss. Following the first 30 days, we observe a drop in pan mass, and thus drying of the salt, when the relative humidity decreases, and a rebound in pan mass when there are increases in the relative humidity.

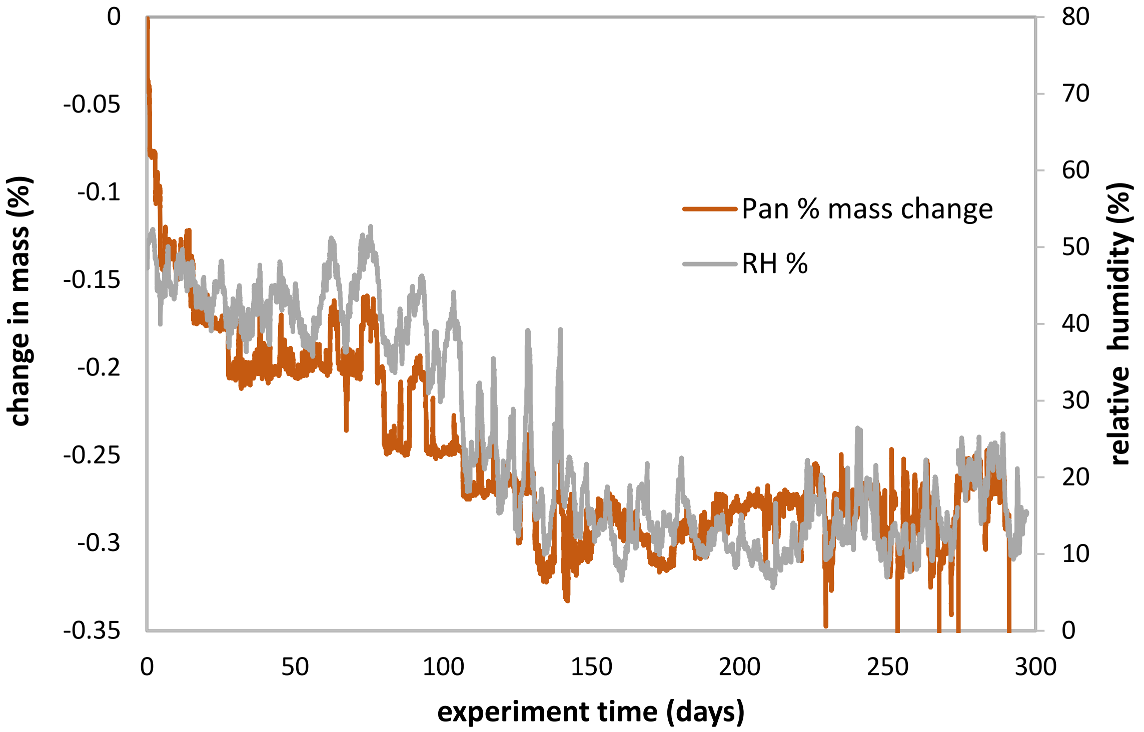

Figure 4: Change in mass (%) of crushed salt and relative humidity of air measured in the underground pan experiment at the WIPP site for the salt pan, for a portion of the experiment run-time between 7/10/2015 and 5/5/2016.

Modeling

Mass changes of the simulated salt cone are compared to measured mass changes in the bench-scale pan experiment for the same time period in Figure 5. Due to uncertainty in the exact dimensions of the salt cone, mass changes were normalized to the percentage change of the total mass. The model results are only reported for an eight-day period (4.5 – 12.5 days); the length of the modeled record was limited by simulation run-time and numerical convergence difficulties. Thus we only observe mass % fluctuations between about 0.1 % and 0.15 % over the 8-day record, a small range when compared to the 12-month experimental record shown in Figure 4 that ranges from 0.0 % to 0.4 %.

Directionality of the modeled changes tracks with measured values, with peak and valley inflection points correlating well. This shows that in situations with airflow with the *fxa* boundary applied, FEHM produces accurate values if humidity, air flow, and initial conditions are well established.

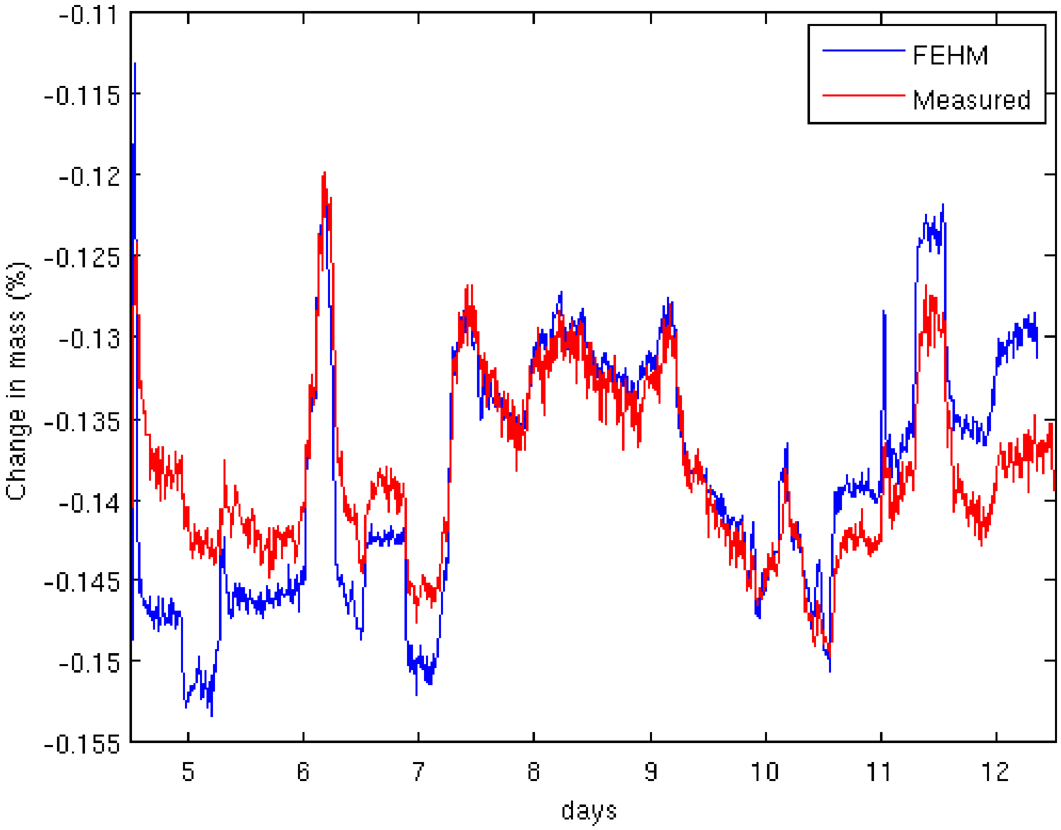

Figure 5: Mass change (%) of salt cone, showing comparison of FEHM model results (blue) to experimental measurements (red).

The results also indicate the vapor pressure lowering regime calculated for RoM salt in the FEHM simulations is appropriate at the WIPP site. Based on field observations, the dissolved WIPP salt and capillary pressure cause the pore water for RoM salt to require a relative humidity of about 25 % than that of pure water. The combination of P_{wv} lowering due to dissolved salt in pore water and with the additional retention resulting from capillary pressure in variably-saturated RoM salt, produces a reasonable match to the experimental data. The salt-dependent P_{wv}-lowering relationship was added to the salt module in FEHM, and tested in this validation study. The good fit between the measured and modeled data suggests the 25 % relative humidity estimate is appropriate for moving forward with more complicated modeling in the future.

CONCLUSIONS

Modeling the complicated and coupled THMC processes around placement of a heat-generating source in variably-saturated, crushed salt includes in many uncertainties. The validation of new FEHM capabilities for simulation of salt and relative humidity by comparison of model results with data from simple experiments, as discussed above, builds confidence in model accuracy. Rates of evaporation and precipitation of water vapor from granular salt depend not only on the relative humidity of air in the subsurface, but also the lowering of the water-vapor pressure for pore water with dissolved salt and capillary retention. The experiment represented a dynamic system with simple constrained parameters which rendered a dataset with which to compare to modeled results. We see a general agreement between the modeled and experimental results, suggesting FEHM is will perform well for simulations including a higher degree of complexity. Thus the newly added salt module is a valuable tool for modeling future experiments and HGNW disposal in bedded salt.

ACKNOWLEDGEMENTS

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REFERENCES

1. McMahon, K. 2012. Update of the Used-Fuel Disposition Campaign Implementation Plan, DOE Office of Nuclear Energy Report FCRD-UFD-2012-000334. Albuquerque, NM.
2. Hansen, F.D., and C.D. Leigh. 2012. Salt disposal of heat-generating nuclear waste. Sandia National Laboratories Report SAND2011-0161. Albuquerque, NM.
3. Kuhlman, K.L., and B. Malama. 2013. Brine Flow in Heated Geologic Salt. Sandia National Laboratories Report SAND2013-1944. Albuquerque, NM.
4. Caporuscio, F.A., H. Boukhalfa, M.C. Cheshire, A.B. Jordan, and M. Ding. 2013. Brine Migration Experimental Studies for Salt Repositories, FCRD Used Fuel Disposition Campaign Milestone FCRD-UFD-2013-000204. Los Alamos, NM.
5. Kelly, W.R. 1985. Brine migration in salt: Topical report. http://pbadupws.nrc.gov/docs/ML0404/ML040410467.pdf (accessed 10 July 2013).
6. Jordan, A.B., H. Boukhalfa, F.A. Caporuscio, B.A. Robinson, P.H. Stauffer. 2015. Hydrous Mineral Dehydration around Heat-Generating Nuclear Waste in Bedded Salt Formations, Environmental Science & Technology, accepted pending revisions.
7. Zyvoloski, G.A., B.A. Robinson, Z.V. Dash, and L.L. Trease. 1997. Summary of the models and methods for the FEHM application—A finite element mass- and heat-transfer code. Los Alamos National Laboratory Report LA-13307-

MS, modified 1999. Los Alamos, NM.

8. Zyvoloski, G.A. 2007. FEHM: A control volume finite element code for simulating subsurface multi-phase multi-fluid heat and mass transfer. Los Alamos National Laboratory Report LA-UR-07-3359. Los Alamos, NM.
9. Kelkar, S., G. WoldeGabriel, and K. Rehfeldt. 2011. Hot Dry Rock Geothermal Energy Development at Los Alamos National Laboratory: 1970-1995, Final Report. Los Alamos National Laboratory Report LA-14433-HDR. Los Alamos, NM.
10. Bourret, S.M., P.J. Johnson, G.A. Zyvoloski, S.P. Chu, D.J. Weaver, S. Otto, H. Bouklhalfa, F.A. Caporuscio, A.B. Jordan, P.H. Stauffer. 2016. Experiments and Modeling in Support of Generic Salt Repository Science. Los Alamos National Laboratory Document, LA-UR-16-27329. Los Alamos, NM.
11. Jordan, A.B., H. Boukhalfa, F.A. Caporuscio, and P.H. Stauffer. 2015. Brine Transport Experiments in Run-of-Mine Salt. Los Alamos National Laboratory Report LA-UR-15-26804. Los Alamos, NM.
12. Jordan, A.B., G.A. Zyvoloski, D.J. Weaver, S. Otto, and P.H. Stauffer. 2015. Coupled Thermal-Hydrologic-Chemical Model for In-Drift Disposal Test. Los Alamos National Laboratory Report LA-UR-15-27442. Los Alamos, NM.
13. Jordan, A.B., P.H. Stauffer, D.T. Reed, H. Boukhalfa, F.A. Caporuscio, and B.A. Robinson. 2014. Draft Test Plan for Brine Migration Experimental Studies in Run-of-Mine Salt Backfill. Los Alamos National Laboratory Report LA-UR-14-27338. Los Alamos, NM.
14. Stauffer, P.H., D.R. Harp, A.B. Jordan, Z. Lu, S. Kelkar, Q. Kang, J. Ten Cate, H. Boukhalfa, Y. Labayed, P.W. Reimus, F.A. Caporuscio, T.A. Miller, and B.A. Robinson. 2013. Coupled model for heat and water transport in a high level waste repository in salt. Los Alamos National Laboratory Document, LA-UR-13-27584. Los Alamos, NM.

   http://www.energy.gov/sites/prod/files/2013/12/f5/CouplModelHeatWaterTransprtGenericHLWRepSalt_1.pdf.
15. Stauffer, P.H., D.R. Harp, and B.A. Robinson (2012). Model Development and Analysis of the Fate and Transport of Water in a Salt-Based Repository, Used Fuel Disposition Campaign Milestone M2FT-12LA08180112, LANL Report LA-UR-12-25050. Los Alamos, NM.
16. Eppelbaum, L.V., I. Kutasov, and A. Pilchin. 2014. Applied Geothermics. Springer Science and Business Media LLC, New York, NY.