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**Analysis of Interactions
of Mechanical Deformations and Mass Transfer
on Heat Transfer from an Underground
Nuclear-Waste Repository**

Technical Report

October, 1979

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ONWI
Office of Nuclear Waste Isolation

BATTELLE Project Management Division

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BIBLIOGRAPHIC DATA

Bloom, S. G., and L. E. Hulbert, 1979. *Analysis of Interactions of Mechanical Deformations and Mass Transfer on Heat Transfer from an Underground Nuclear Waste Repository*, ONWI-64, Office of Nuclear Waste Isolation, Battelle Memorial Institute, Columbus, OH.

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Printed in the United States of America
Available from
National Technical Information Service
U.S. Department of Commerce
5285 Port Royal Road
Springfield, VA 22161

NTIS price codes
Printed copy: A02
Microfiche copy: A01

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This report was prepared by Battelle-Columbus Laboratories under Subcontract E516-01900 with Battelle Project Management Division Office of Nuclear Waste Isolation under Contract No. DE-AC06-76RLO1830-ONWI with the U.S. Department of Energy. This subcontract was administered by the Battelle Office of Nuclear Waste Isolation.

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FINAL REPORT
on
ONWI-64
ANALYSIS OF INTERACTIONS
OF MECHANICAL DEFORMATIONS AND MASS TRANSFER
ON HEAT TRANSFER FROM AN UNDERGROUND
NUCLEAR WASTE REPOSITORY

to
BATTELLE PROJECT MANAGEMENT DIVISION
OFFICE OF NUCLEAR WASTE ISOLATION

by
S. G. Bloom and L. E. Hulbert

BATTELLE
Columbus Laboratories

October, 1979

SUMMARY

This research program consisted of five tasks: (1) review existing models of heat transfer and thermal stress in geologic materials, (2) develop mathematical formulations for mechanisms of interactions that are missing from existing models, (3) examine the magnitude of these interactions using simplified analytical procedures, (4) develop approximate methods and models to simulate significant interactions, and (5) recommend any additional efforts that might be appropriate to better quantify the significant interactions.

A review of existing models identified several effects that may need consideration in further model development. Most of these effects involved coupling equations through variable property values rather than through omission of any significant mechanism. However, it was also shown that more than one mechanism may adequately simulate a given set of experimental data and additional experimental data are needed to establish which (if any) of the possible mechanisms would actually control conditions in a nuclear waste repository. In particular, it is believed that mathematical modeling of major thermomechanical effects can be accomplished with finite element analysis computer programs, provided that adequate thermomechanical property data of salt and overburden are attained.

An attempt was made to develop a general set of differential equations for simulating momentum, mass, and energy flows in geologic formations in order to illustrate the possible mechanisms and point out those included and not included in existing models. Most of the mechanisms are included in some manner in existing models although some approximations may not be adequate. More experimental data are required to assess the importance of most omitted mechanisms.

Analysis of some data on brine migration in salt indicated that two mechanisms, acting simultaneously, could adequately explain the flow. These are Darcy flow and a combination of ordinary and thermal diffusion enhanced by temperature-dependent solubility. Equations based on this simultaneous action correlated the data very well and indicated the possible need to include both (and, maybe other) mechanisms in future models. A program is recommended for further study of brine mobility. An expected result of this program includes recommendations for further experimental work.

INTRODUCTION

The heat released from nuclear wastes stored in an underground repository will produce temperature elevations in the surrounding geologic materials. Heat transfer analyses which attempt to simulate these temperature elevations may not include the interactions of the heat transfer with mechanical deformations of the geologic materials and mass transfer in these materials. The magnitudes of these effects need not be large in order to produce significant errors in the simulated temperatures over the long periods of time which must be considered in nuclear waste repositories. The inclusion of these interactions would increase the complexity of the heat transfer analyses and would limit the utility of the resulting computer codes. It is therefore desirable to first use simplified analytical procedures in order to examine the magnitude of these interactions under conditions that might be encountered around a nuclear waste repository. With such procedures, an assessment could be made as to which, if

any, interactions might have a significant effect on the simulated temperatures. Then, only those which may have a significant effect need be included in more detailed heat transfer analyses.

The purpose of this research program was to identify those interactions which may significantly affect the heat transfer around the repository. To the extent possible, approximate methods and/or models were to be developed for quantitatively estimating these effects. The research program consisted of five tasks: (1) review of existing models of heat transfer in geologic materials, (2) develop mathematical formulations for mechanisms of interactions that are missing from existing models, (3) examine the magnitude of these interactions using simplified analytical procedures, (4) develop approximate methods and models to simulate significant interactions, and (5) recommend any additional efforts that might be appropriate to better quantify the significant interactions.

REVIEW OF EXISTING MODELS OF HEAT TRANSFER IN GEOLOGIC MATERIALS

This review of existing models was conducted during the first 3 months of the program and included a review of various documents plus visits to some of the principal contractors involved in developing mathematical models of nuclear waste repositories. Visits were made to the following organizations:

- (a) RE/SPEC, Rapid City, South Dakota
- (b) SAI, San Francisco, California
- (c) University of Minnesota, Minneapolis, Minnesota
- (d) Lawrence Livermore Laboratories, Livermore, California
- (e) ORNL, Oak Ridge, Tennessee.

With the exception of the group at Livermore (who were working on an NRC contract), all of the groups visited were contractors on ONWI programs. In each case, the personnel were open and candid about discussing the facets of their research. A number of useful references were also identified as a result of these visits. These were collected and studied. Table 1 is a list of the references which were most often used during this research program.

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Effects Considered in Existing Models

The review revealed the existence of the following general classes of mathematical models applied to nuclear waste repositories: (1) heat transfer, (2) thermomechanical response, (3) groundwater hydrology and radionuclide transport, and (4) brine migration in salt. Most of the heat transfer models include temperature and geometric (direction and position) variations in the properties of the medium and can handle almost any consistent set of initial and boundary conditions. None presently includes property variations as a function of stress or changes in the composition of the medium that might result from some transport mechanism (e.g., groundwater flow or transport induced by the thermal gradient). The models generally use either finite-difference or finite-element numerical procedures.

A great deal of effort has been placed on developing thermo-mechanical models. In most of the existing models, the mechanical stresses and deformations are dependent on temperature. With one exception, effects of stress or deformation are ignored in computing temperatures. (There are models which include convective heat transfer in groundwater flow. Strictly speaking, these could be considered as heat transfer effected by the mechanical state). In thermomechanical models of repositories, the constitutive properties can be functions of time, temperature, stress level, and geometry. Variations with respect to composition are not presently considered. Existing computer models can handle almost any consistent set of initial and boundary conditions. These models use finite-element techniques exclusively for the stress analysis. Some investigators combine a finite-difference heat transfer portion with a finite-element mechanics portion.

One interesting approach is to model the overburden as an elastic continuum. The near-field nonlinear creep deformations of the repository are input as boundary conditions on the overburden model. This permits repository closure calculations to be carried out with greatly reduced computer times.

Existing models of groundwater hydrology and radionuclide transport generally include the effect of overburden pressure on the properties governing the water flow (i.e., permeability) but do not include effects of stresses induced by temperature or by the water flow itself. (However, there are models which simulate the stresses and mechanical deformations induced by the water flow.) The properties can be functions of geometry but effects of temperature and changes in composition of the medium are not considered. Radionuclide transport is simulated by assuming that instantaneous equilibrium exists between the radionuclide in water and the radionuclide attached to the medium. The equilibrium distribution coefficient is therefore a key parameter and many models consider its variation as a function of chemical composition of the water. Variations as a function of temperature and stress are not presently included. Most of these models use finite-element or finite-difference techniques although some of the simpler ones use analytical techniques.

Brine migration under the influence of a temperature gradient in salt is driven by several mechanisms which include (a) thermal diffusion in the liquid, (b) ordinary diffusion in the liquid and gas, (c) temperature-dependent solubility in the liquid, (d) temperature- and concentration-dependent vapor pressure of the liquid, and (e) surface energy at crystal dislocations. Calculations have been made with relatively simple models which consider constant-property, thermal and ordinary diffusion in the liquid, plus temperature-dependent solubility in the liquid. Differential equations are available which include all of the above mechanisms but there are no existing models for solving them. An alternative mechanism proposed for brine migration is based on Darcy flow (flow through a porous material), in which the driving force is the difference between the vapor pressure of water over the brine and the confining pressure. As shown later in this report, this mechanism provides a plausible means for correlating some data on brine migration.

Effects Which May Need Consideration

A major objective of this study was to identify effects which are not included in existing models and which may need more consideration in future model development. The effects discussed in the following paragraphs were identified on the basis of the literature review, discussions with the other contractors, and the experience of the authors of this report.

In considering effects omitted from existing models, a distinction should be made between effects caused by omission of a significant mechanism versus effects caused by neglecting property variations. In an initial review of existing models several effects were identified which may need consideration in further model development. All of these effects involved coupling equations through variable property values rather than through omission of any significant mechanism. However, further consideration and examination of some data on brine migration have emphasized that more than one mechanism may adequately simulate a given set of experimental data. In many of these cases, further experimental data are needed to establish which (if any) of the possible mechanisms would actually control conditions in a nuclear waste repository. Inclusion of all of these possibilities in a model would require additional complexity but would not necessarily increase the precision of the models without better accuracy of property values. Consequently, existing models generally focus on one of the possible mechanisms and ignore the others.

The following discussion emphasizes those effects involving coupling through variable property values. The possibility of alternative mechanisms is discussed later in this report and is based on (1) the development and examination of a general set of differential equations for simulating momentum, mass, and energy flows in geologic formations, and (2) analyses of brine migration data.

Groundwater Flow Coupled With Heat Transfer

The convective heat flow resulting from groundwater flow and the temperature variation of the properties of water are presently being modeled. However, there are a number of other interactions which may be significant. The solubility of various constituents in the water (including any radionuclides leaking from the waste canisters) could be a function of temperature and could affect their transport rate. If heat-generating radionuclides are being transported with the water, the solubility difference could affect the radionuclide distribution in the medium which, in turn, would affect the geometry of the heat source. The flow-related properties (permeability and porosity) of the medium may be temperature dependent.

Groundwater Flow Coupled With Stresses

Models exist for calculating stresses resulting from groundwater flow and it is known that permeability and porosity, two key parameters in simulating groundwater flow, vary with stress. The effect of temperature- and flow-induced stresses is therefore likely to be very significant and may only be discernible if the flow and mechanics models are coupled. In addition, the composition changes accompanying the groundwater flow could lead to variations in the constitutive properties of the medium which could affect the stress levels.

Coupling Between Heat Transfer and Thermomechanical Response

Coupling between the heat transfer and stress phenomena could occur in one of two ways: (1) through geometrically nonlinear large deformations, and (2) through nonlinear material property effects.

With one exception, most thermomechanical response analyses have indicated that the thermomechanical response of repositories is within the geometric range which permits uncoupling thermal and stress analyses. The exception involves very-near-field nonlinear creep deformation around a cask for which the geometry change is enough to affect the heat transfer analysis.

Material property coupling effects may include pore closure or opening in salt as a result of the thermal strains or the opening or closing of fissures in the rock overburden.

Heat Transfer Coupled to Mechanics

Present models do not consider the effects of stresses on heat transfer properties but thermal conductivity and heat capacity may vary with stress. This may be especially true with viscoelastic materials, in which the deformations are large enough to close pores and alter directional differences in conductivity.

Complete Coupling of Groundwater Flow With Heat Transfer and Mechanics

It may be necessary to couple all three mechanisms for an adequate simulation. The effects caused by two-way coupling could also interact in a three-way coupling.

Brine Migration Coupled With Heat Transfer and Mechanics in Salt

Brine migration under the influence of a temperature rise in salt has been shown to occur but the amount of mass transfer is not considered large. Therefore, it is not likely that this mechanism alone could lead to significant releases of radioactivity to the environment. However, the brine migration could significantly alter the heat transfer and mechanical properties of the salt, which could lead to alterations of the temperature gradient, stress-induced migration, and parting at crystal dislocations.

GENERAL EQUATIONS FOR SIMULATING MOMENTUM, MASS,
AND ENERGY FLOWS IN GEOLOGIC FORMATIONS

An attempt has been made to develop a general set of differential equations for simulating momentum, mass, and energy flows in geologic formations in order to illustrate the possible mechanisms and point out those included and not included in existing models.

It is believed that mathematical formulations have been developed that could represent the important thermomechanical effects in repositories. These include both the nonlinear material behavior and the nonlinear effects of near-field large deformations. Detailed representation of material behavior must wait for the measurements of the appropriate properties. Development of the approach to modeling large strains and large deformations will depend on the needs for modeling the repository in the short and long term.

Momentum Flow

It is generally acknowledged that momentum flow in geologic formations leads to low fluid velocities. In the presence of a pressure gradient or buoyant forces, the momentum flow reduces to Darcy's law for velocity which is

$$U_i = (k_i/\mu_i) \nabla (p_i + \rho_i gh), \quad (1)$$

where U_i is the velocity of Fluid i, cm/sec
 k_i is the formation permeability to Fluid i
in the presence of all other fluids, cm^2
 μ_i is the viscosity of Fluid i, $\text{g}/(\text{cm sec})$
 p_i is the pressure of Fluid i, dynes/cm^2
 g is the acceleration of gravity, cm/sec^2
 h is the height above some reference plane, cm
 ρ_i is the density of Fluid i, g/cm^3 .

Each fluid is a different phase (i.e., gas and/or immiscible liquids) in the formation and Equation (1) applies to each phase.

Mass Flow

The total mass flow in a geologic formation is given by

$$\partial(\phi \rho_i S_i)/\partial t = -\nabla \cdot (\rho_i U_i), \quad (2)$$

where ϕ is the formation porosity

S_i is the fraction of pore spaces occupied by Fluid i (saturation)

t is time, sec.

The mass flow for each component is

$$\begin{aligned} \partial(\phi \rho_i S_i C_{ni})/\partial t = & -\nabla \cdot (\rho_i U_i C_{ni}) + \nabla \cdot (D_{ni} \nabla [\rho_i C_{ni}]) \\ & -\nabla \cdot (D_{ni} \sigma_{ni} C_{ni} \rho_i \nabla T_i) + \phi S_i \sum_j F_{nji} R_{ji} \\ & + \sum_{m \neq i} K_{nim} (C_{Enim} - C_{ni}), \end{aligned} \quad (3)$$

where C_{ni} is the mass fraction of Component n in Fluid i

D_{ni} is the ordinary diffusivity of Component n in Fluid i, cm^2/sec

T_i is the temperature of Fluid i, C

σ_{ni} is a ratio of thermal to ordinary diffusivity (Soret coefficient), $(\text{C})^{-1}$

R_{ji} is the rate of Chemical Reaction j in Fluid i, g limiting reactant/ $(\text{cm}^3 \text{ sec})$

F_{nji} is the mass of Component n produced per unit mass of limiting reactant in Reaction j.

C_{Enim} is the mass fraction of Component n in Fluid i which would be in equilibrium with the mass fraction of Component n in Fluid m

K_{nim} is the mass transfer coefficient for Component n from Fluid m to Fluid i, $\text{g}/(\text{cm}^3 \text{ sec})$.

As indicated by the definitions of the parameters, there are five mechanisms which are believed to control mass transfer in a nuclear waste repository and these are the five terms on the right hand side of Equation (3). In order, these are convection, ordinary diffusion, thermal diffusion, chemical reactions, and interphase mass transfer. The interphase mass transfer term also includes the possibility of mass transfer from the formation medium (solid) to Fluid i.

Energy Flow

The equation for energy flow can be written in terms of a total energy defined by

$$E_i = H_i + gh, \quad (4)$$

where E_i is the total energy of Fluid i, J/g

H_i is the enthalpy of Fluid i, J/g.

Enthalpy is found from the relations

$$H_i = \sum_n H_{ni} C_{ni}, \quad (5)$$

where H_{ni} is the specific enthalpy of Component n in Fluid i at the conditions of Fluid i.

Specific enthalpy is related to temperature by

$$H_{ni} = H_{oni} + \int_{T_o}^T c_{nid} T, \quad (6)$$

where H_{oni} is the specific enthalpy of Component n in Fluid i at some reference temperature, T_o , J/g

c_{ni} is the constant pressure heat capacity of Component n in Fluid i, J/(gc).

The equation for the total energy flow is

$$\begin{aligned} \partial(\phi \rho_i S_i E_i) / \partial t = & -\nabla \cdot (\rho_i E_i U_i) + \partial(\phi S_i p_i) / \partial t \\ & + \nabla \cdot (\kappa_i \nabla T_i) + \phi S_i \sum_j R_{ji} H_{Rj} + \sum_{m \neq i} h_{im} (T_m - T_i) \end{aligned} \quad (7)$$

where κ_i is the thermal conductivity of Fluid i, J/(cm C sec)

H_{Rj} is the energy released during Reaction j, J/g limiting reactant

h_{im} is the heat transfer coefficient from Fluid m to Fluid i, J/(cm³ C sec)

T_m is the temperature of Fluid m, C.

The mechanisms for energy flow included as the right hand side of Equation (7) are, in order, convection, pressure-volume work, conduction, heat of chemical reactions, and interphase heat transfer. Viscous dissipation is assumed negligible for a slow-flowing fluid in the presence of relatively large temperature gradients. Also neglected in Equation (7) is nuclear energy (gamma radiation) which may affect material properties but,

because of shielding, it is assumed negligible in comparison to the heat flow. The interphase heat transfer term also includes the possibility of transferring heat from the formation medium to Fluid i.

Terms Omitted From Existing Models

Momentum flow and mass flow are combined in existing models to simulate single phase (groundwater) flow and radionuclide transport in ground water. These models are equivalent to a combination of Equations (1), (2), and (3), in which the mechanisms for thermal diffusion and chemical reaction are ignored. Interphase mass transfer is often approximated by assuming instantaneous equilibrium exists between the radionuclide in ground water and the radionuclide attached to the formation. Chemical reaction is sometimes approximated by modifying the equilibrium constant used to approximate interphase mass transfers. Thermal diffusion is probably not significant for these cases but the assumption of only a single fluid phase may not be completely accurate. The brine migration experiment, described later in this report, implies the possibility of a vapor phase under some conditions.

Two types of existing models simulate brine flow in salt. One model is based on Equation (3) only and assumes the flow occurs as a single fluid (brine) under the influence of ordinary and thermal diffusion. The other model is based on a combination of Equations (1) and (2) and assumes the presence of two phases (brine and water vapor). Both models have been reasonably successful in correlating some of the existing data on brine flow but more data are needed to verify which is more correct.

Existing models for energy flow are almost exclusively heat transfer models. Reactions and pressure-volume work are neglected. Convection and interphase heat transfer are included when considering groundwater flow in the immediate vicinity of the nuclear waste canisters. Inclusion of all these mechanisms is not expected to increase the precision of the existing models without better accuracy of material property data.

ANALYSIS OF SOME DATA ON BRINE MIGRATION IN SALT

One of the models for brine migration in salt attributes the migration principally to a combination of ordinary diffusion enhanced by the temperature-dependent solubility of salt in water, and thermal diffusion. These mechanisms lead to brine migration up the thermal gradient if there is no vapor space in the brine inclusions, and down the gradient if there is vapor. However, an experiment at ORNL indicates significant migration can occur both up and down the gradient. Another model is based on Darcy flow in which the driving force for flow is a pressure gradient. For the ORNL experiment, the gradient is the difference between the vapor pressure of water over the brine within the salt, minus the confining pressure on the salt. The temperature of the entire salt specimen in the ORNL experiment was high enough so that the vapor pressure exceeded the confining pressure at both the hot and cold sides. Under these conditions, brine would escape from both sides according to a Darcy flow model. This, in fact, was observed in the ORNL experiment.

Brine Migration Experiments

The essential features of the ORNL experiment are shown in Figure 1. A cylinder of halite, with radius r_0 , was subjected to a steady-state temperature gradient as shown in Figure 1a. The cold side ($z=0$) was at 105 C, while the hot side ($z=z_3$) was 190 C. The region between z_1 and z_2 is assumed to be saturated with brine, while the rest of the cylinder is assumed to contain vapor. The vapor pressure of water over the brine within the cylinder, p_v , would follow a function similar to that shown in Figure 1b, and range from p_1 at the cold side to p_2 at the hot side. If brine migration is due to Darcy flow, flow would occur from both faces if the confining pressure p_0 , is less than p_1 . If the confining pressure is between p_1 and p_2 , flow will only occur from the hot side, while no flow occurs if the confining pressure exceeds p_2 . In the ORNL experiment, p_0 was less than p_1 and Darcy flow could occur from both sides.

1a. Halite Cylinder

1b. Pressure

FIGURE 1. ORNL HALITE CYLINDER EXPERIMENT

Another type of experiment was conducted at Sandia Laboratories and is shown in Figure 2. A cylindrical block of salt was heated in the center and cooled at its circumference. The top and bottom of the cylinder were insulated. Brine flowing into the centrally heated area was collected but no brine measurements were reported from the outer circumference. Either there was no flow from this cooled area or there were simply no measurements of the flow.

Mathematical Analysis of the Brine Migration Experiments

Both the ORNL and Sandia experiments had a period during which the heat flow was assumed to be at a steady-state condition. For the ORNL experiment, this occurred after about 5 days through 22 days, while for the Sandia experiment, the period was about 45 days through 120 days. Only these steady-state periods were considered in this analysis. The end temperatures were fixed in the ORNL experiment and heat and mass flow were confined to the axial direction. The temperature gradient is therefore given by $(T_3 - T_0)/z_3 = (190 - 105)/7.0 = 12.14 \text{ C/cm}$. In the Sandia experiment, the temperature at the outer circumference was maintained at about 21 C, while the heater output was maintained at 1500 watts. If this heat is assumed to have been uniformly distributed over the entire length (1.0 m) of the inner radius instead of the actual heater length (0.6 m), the temperature gradient is a simple function of radius and is given by

$$dT/dr = q/(2\pi Lr\kappa), \quad (8)$$

where r is the radial distance, cm

T is the temperature, C

q is the heating rate, 1500 J/sec

L is the heated length, taken as the entire 100 cm length of the block

κ is the thermal conductivity of the solid salt, about 0.043 J/cm sec C.

With the outer circumference set at about 21 C, the inner radius is about 150 C.

FIGURE 2. SANDIA SALT CYLINDER

An expression for brine migration attributable to Darcy flow may be derived by assuming the salt can be divided into two regions; one containing only brine ($S_1=1.0$) and one containing only vapor ($S_2=1.0$). The brine is assumed to be virtually immobile and Equation (2) only applies to the vapor region. The boundary between the two regions is a surface which can be described by a function, $F=0$. A point on this surface will move normal to the surface, and rate of motion is determined by equating the mass of vapor lost from an infinitesimal volume next to the boundary surface, to the mass of brine lost as a result of the motion of the surface. This leads to

$$V = dF/dt = [(k_2 \rho_2 / (\phi \rho_1 \mu_2)) \nabla p \cdot \nabla F] [\nabla F / (\nabla F \cdot \nabla F)], \quad (9)$$

where $F=0$ is the locus of points on the boundary surface

V is the velocity of a point on the surface, cm/sec

Subscript 1 refers to brine and 2 refers to water vapor.

The pressure gradient in Equation (9), ∇p , is determined from the solution of Equation (2) in the vapor region ($S_1=0$ and $S_2=1.0$) with $p=p_v$ at the boundary surface ($F=0$).

The boundary between the two regions generally moves slowly in comparison to the time it takes the vapor to set up a steady-state pressure profile. This allows us to assume a continuously-linear pressure profile in the vapor region and to write the following expressions for the cumulative brine flow from the ORNL experiment as a result of Darcy flow:

$$dW_o/dt = \pi^2 r_o^4 \phi \rho_1 k_2 \rho_2 [p_v(z_1) - p_o] / (W_o \mu_2) \quad (10)$$

$$dW_3/dt = \pi^2 r_o^4 \phi \rho_1 k_2 \rho_2 [p_v(z_2) - p_o] / (W_3 \mu_2), \quad (11)$$

where W_o is the cumulative brine flow from the cold side ($z=0$) of the ORNL halite cylinder, g

W_3 is the cumulative brine flow from the hot side ($z=z_3=7.0$ cm) of the ORNL cylinder, g

$p_v(z)$ is the vapor pressure at axial distance z , dyne/cm²

p_o is the confining pressure on the salt, dyne/cm².

The corresponding expression for brine flow from the Sandia experiment (radial flow) is

$$dW_c/dt = 2\pi L k_2 \rho_2 [p_v(r_1) - p_o] [\mu_2 \ln(r_1/r_H)]^{-1}, \quad (12)$$

where W_c is the cumulative brine flow from the heated hole in the salt cylinder, g
 r_1 is the radial position of the boundary between the brine and vapor, cm
 r_H is the radius of the heated hole, 5 cm.

Expressions can also be written for cumulative brine flow when the flow is caused by thermal and ordinary diffusion enhanced by the temperature-dependent solubility of salt in brine. The expression applicable to the ORNL experiment is

$$dW_3/dt = 1.4\pi r_o^2 \phi_1 (C_{S1}/C_{S4}) D_{S1} [(\partial C_{ES1,4}/\partial T)/C_{S1} - \sigma_{S1}] (T_3 - T_o) z_3, \quad (13)$$

where Subscript 4 refers to solid salt

Subscript S refers to salt as a component in brine.

The mechanisms in Equation (13) cannot account for flow from the cold side in the ORNL experiment and would subtract from any cold-side Darcy flow if both mechanisms operated. The corresponding expression for the Sandia experiment is

$$dW_c/dt = 1.4\phi_1 q (C_{S1}/C_{S4}) D_{S1} [(\partial C_{ES1,4}/\partial T)/C_{S1} - \sigma_{S1}] / \kappa. \quad (14)$$

In general, the density and viscosity of the vapor, the porosity and permeability of the salt, and the solubility and diffusivity ratios of the brine are functions of temperature and pressure. However, if the temperature gradient is not too steep, these properties may be assumed constant. Also, the vapor pressure is generally a nonlinear function of temperature but, for small temperature changes, may be assumed linear with temperature. These assumptions were not actually met in the ORNL and Sandia experiments. However, using these assumptions leads to analytical solutions of the equations which can be easily manipulated in order to examine some of the flow characteristics. Without these assumptions, numerical solutions are necessary.

With the above assumptions, the solutions for combined Darcy and thermal diffusion flow in the ORNL experiment are

$$W_o - \alpha \ln(1 + W_o/\alpha) = \beta t - \beta t_o \quad (15)$$

$$W_3 + (\alpha + W_i) \ln[1 - W_3/(\alpha + W_i)] = -\beta t + \beta t_o, \quad (16)$$

where

$$E = 1.4 \phi \rho_1 \mu_2 (T_3 - T_o) (C_{S1}/C_{S4}) D_{S1} [(\partial C_{ES1,4}/\partial T)/C_{S1} - \sigma_{S1}] [k_2 \rho_2 (p_v(z_3) - p_v(o))]^{-1}$$

$$\alpha = \pi r_o^2 \phi \rho_1 z_3 (p_v(o) - p_o) [(p_v(z_3) - p_v(o))(1-E)]^{-1}$$

$$W_i = \pi r_o^2 \phi \rho_1 z_3 / (1-E)$$

$$\beta = \pi r_o^2 k_2 \rho_2 [p_v(z_3) - p_v(o)] (1-E) / (z_3 \mu_2)$$

$p_v(o)$ is the vapor pressure at the cold side ($z=0$), dynes/cm².

Note that the thermal diffusion adds to the flow from the hot side but subtracts from the flow from the cold side. The corresponding solution for brine flow in the Sandia experiment is

$$-W_c + a W_H \exp(a) [E_1(a - \ln(W_c/W_H + 1)) - E_1(a)] = bt - bt_o, \quad (17)$$

where

$$\delta = 1.4 \phi \rho_1 \mu_2 (C_{S1}/C_{S4}) (T_H - T_c) D_{S1} [(\partial C_{ES1,4}/\partial T) C_{S1} - \sigma_{S1}] [k_2 \rho_2 (p_v(r_H) - p_v(r_c))]^{-1}$$

$$a = 4\pi L \kappa (p_v(r_H) - p_o) (T_H - T_c) [q(1-\delta) (p_v(r_H) - p_v(r_c))]^{-1}$$

$$b = k_2 \rho_2 q [p_v(r_H) - p_v(r_c)] (1-\delta) [\mu_2 \kappa (T_H - T_c)]^{-1}$$

$$W_H = \pi r_H^2 \phi \rho_1 L$$

$$E_1(a) = \int_a^\infty \exp(-t) dt/t$$

r_H is the radius of the heated hole, 5 cm
 r_c is the radius of the cooled, outer circumference, 50 cm
 T_H is the temperature at r_H , 150 C
 T_c is the temperature at r_c , 21 C
 E_1 is the exponential integral function.

The terms E and δ in the the above expressions are measures of the ratio of thermal diffusion to Darcy flow. If these terms are zero, the flow is entirely attributable to Darcy flow.

The above expressions were fitted to the corresponding data from the ORNL and Sandia experiments and the results are shown in Figures 3 and 4. The fitted results are the smooth, solid curves in the figures, while the circles are the measured data points. The cold side data in Figure 3 fit the expression

FIGURE 3. DATA FROM THE ORNL HALITE CYLINDER EXPERIMENT

FIGURE 4. DATA FROM THE SANDIA EXPERIMENT

$$t = 6.77 + 163.9 W_o - 11.05 \ln (1+W_o/0.0059) \quad . \quad (18)$$

The hot side data fit the expression

$$t = 4.08 - 497.7 W_3 - 622.1 \ln (1-W_3/1.23) \quad . \quad (19)$$

The Sandia data in Figure 4 fit the expression

$$t = 45.4 - 5.896 W_c + 32839 [E_1(3.6329 - \ln(1 + W_c/40.537)) - E_1(3.6329)] \quad . \quad (20)$$

The time, t , in Equations (18), (19), and (20) is expressed in days, while the cumulative brine flow is in grams.

The coefficients in Equations (18), (19), and (20) can be used to estimate the porosity (ϕ) and permeability (k_2) of the salt cylinders in the experiments if data are available for the other parameters. The confining pressure in each experiment (p_o) was assumed to be 1 atm or about 1.0×10^6 dynes/cm². The vapor pressures (p_v) were assumed to be identical to pure water and are (in dynes/cm²) about 2.5×10^4 at 21 C, 1.2×10^6 at 105 C, 4.8×10^6 at 150 C, and 1.3×10^7 at 190 C. Estimates of other data are $\rho_2 \approx 0.00059$ g/cm³, $\mu_2 \approx 0.00015$ g/cm sec, $\rho_1 \approx 1.00$ g/cm³, $(C_{S1}/C_{S4}) \approx 0.07$, $D_{S1} \approx 0.0001$ cm²/sec, $(\partial C_{ES1,4}/\partial T)/C_{S1} \approx 0.004/C$, and $\sigma_{S1} \approx -0.002/C$. With these data estimates, the cold side data of the ORNL experiment yield values of 0.0009 for porosity and 2×10^{-16} cm² for permeability, while the hot side data yield 0.0003 for porosity and 6×10^{-17} cm² for permeability. The ratio of thermal diffusion to Darcy flow (E) is about 0.65 from both sets of data. The corresponding values from the Sandia experiment are 0.005 for porosity, 6×10^{-17} cm² for permeability and 0.84 as the ratio of thermal diffusion to Darcy flow (δ). The permeability values are in relatively good agreement and indicate a very tight porous medium which would be expected for salt. The porosity values show considerable variation between data sets, and only the value derived from the Sandia experiment is in good agreement with porosity estimates derived from measured brine content (about 0.004). These variations may be attributed to the effects of differential thermal stresses which can alter the size of pores.

CONCLUSIONS AND RECOMMENDATIONS

A review of existing efforts to mathematically model heat transfer from an underground nuclear waste repository indicates that most of

the expected interactions of mechanical deformations and mass transfer occur through variable material property values. However, there appear to be instances where more than one mechanism may adequately correlate existing data and the existing models do not include all these possibilities. A set of generalized differential equations has been presented to indicate some of these missing mechanisms and a relatively detailed analysis of data on brine migration has shown how a combination of mechanisms can correlate the data. It therefore seems appropriate to examine brine mobility phenomena more closely in order to better understand alternative mechanisms for this flow and to suggest further experimental work that may help to establish the relative magnitude of the mechanisms and their significance in the operation of a nuclear waste repository.

A program is recommended in which existing data on brine mobility are examined in order to test their correlation with mechanisms already proposed, as well as with alternative mechanisms. Suitable mathematical formulations would be developed for all these mechanisms. It is likely that more data will be needed in order to determine which mechanism is dominant or whether more than one mechanism is required to adequately simulate brine mobility. The program would therefore generate recommendations for further experiments to obtain the required data.

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