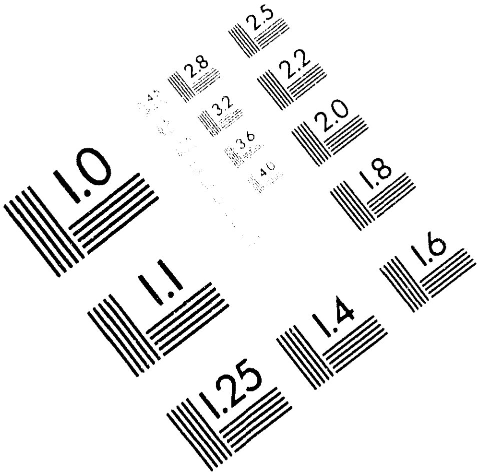

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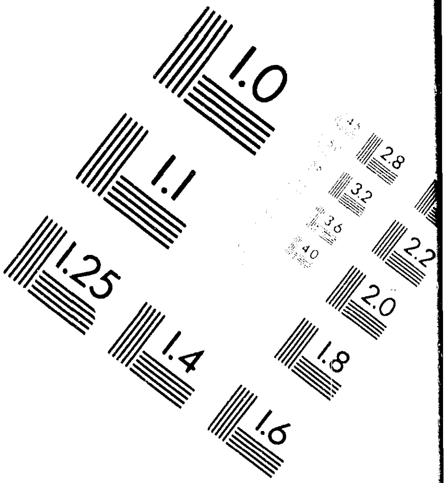

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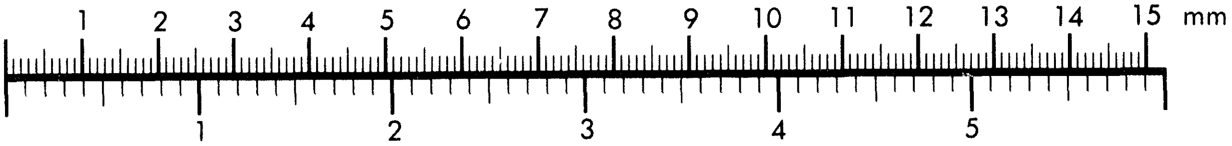

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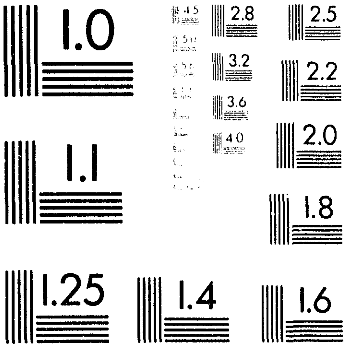

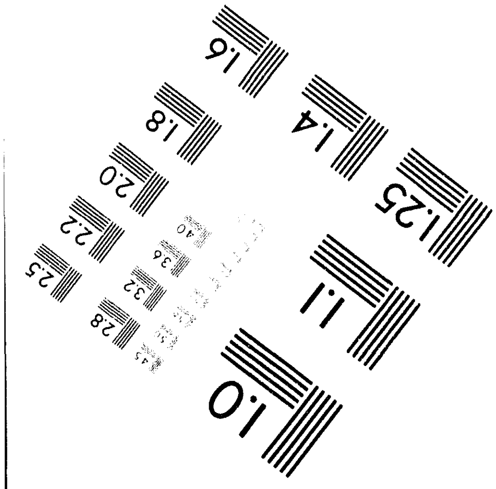

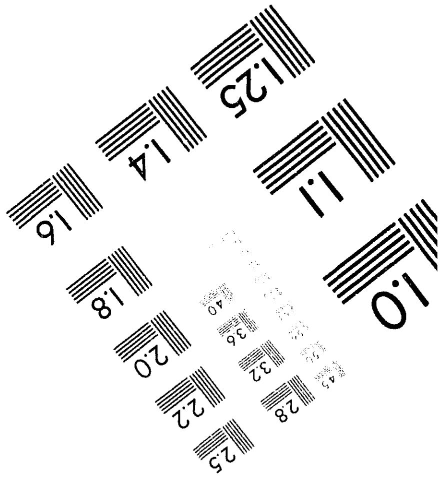

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Phenomenological Studies of Two-Phase Flow Processes for Nuclear Waste Isolation

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PHENOMENOLOGICAL STUDIES OF TWO-PHASE FLOW PROCESSES FOR NUCLEAR WASTE ISOLATION

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ABSTRACT

The U.S. civilian radioactive waste management program is unique in its focus on a site in the unsaturated zone, at Yucca Mountain, Nevada. Two-phase flow phenomena can also play an important role in repositories beneath the water table where gas is generated by corrosion, hydrolysis, and biological degradation of the waste packages. An integrated program has been initiated to enhance our understanding of two-phase flow behavior in fractured rock masses. The studies include two-phase (gas-liquid) flow experiments in laboratory specimens of natural rock fractures, analysis and modeling of heterogeneity and instability effects in two-phase flow, and design and interpretation of field experiments by means of numerical simulation. We present results that identify important aspects of two-phase flow behavior on different space and time scales which are relevant to nuclear waste disposal in both unsaturated and saturated formations.

I. INTRODUCTION

Two-phase flow processes may affect the performance of nuclear waste repositories in a variety of hydrogeologic settings. The relevance of two-phase (gas-liquid) flow is obvious in the Yucca Mountain Project (YMP) which focuses on a site in the unsaturated zone. The Swiss National Cooperative for the Disposal of Radioactive Waste (NAGRA) is interested in two-phase flow phenomena due to the fact that a significant amount of gas may be generated by corrosion, hydrolysis, and biological degradation of the waste packages.[1] For a repository beneath the water table, this may lead to the development of a highly pressurized unsaturated zone around the caverns.[2] Research on two-phase flow in fractured rock masses is among the tasks being jointly pursued by the U.S. DOE and NAGRA under the terms of a cooperative project agreement. A number of phenomenological studies has been conducted to enhance the understanding of two-phase flow processes relevant to nuclear waste isolation. Our current efforts include laboratory experiments using transparent fracture replicas and actual rock fractures. Flow of water and gas under controlled pressure and flow conditions was visualized and measured, and relative permeability curves for single fractures were obtained. Theoretical studies and numerical experiments were conducted to analyze water infiltration in heterogeneous media. Ventilation and tracer experiments probing two-phase flow and transport are currently underway at the Grimsel Test Site (GTS), an underground laboratory operated by NAGRA. Design calculations using LBL's multiphase code TOUGH2[3] have been performed based on an inverse modeling analysis of similar ventilation tests previously conducted at that site.

II. LABORATORY FRACTURE FLOW EXPERIMENTS

We have developed a laboratory apparatus for controlled two-phase flow experiments (gas-liquid) in rough-walled fractures (see Fig. 1).[4, 5] Transparent replicas were fabricated from actual fracture specimen from different field sites, to allow two-phase flow behavior to be visualized. Specially designed endcaps provide control of liquid and gas pressures separately and individually, so that flow behavior can be observed at controlled capillary pressure conditions. All parameters entering into a multi-phase extension of Darcy's law are either controlled or measured, providing a direct means of obtaining relative permeabilities and capillary pressures.

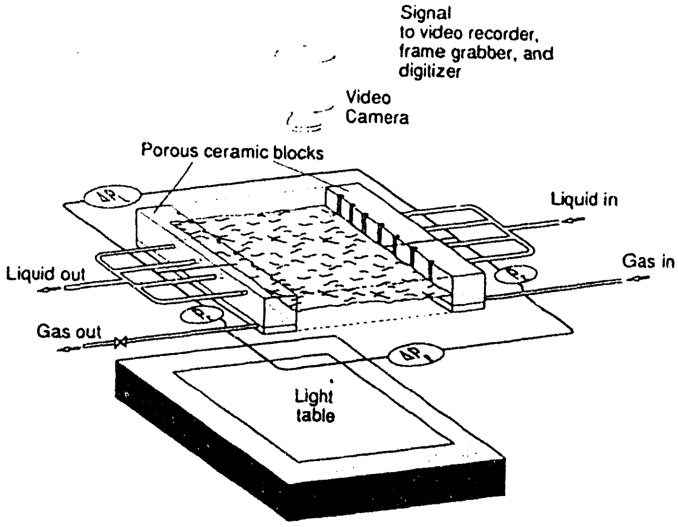

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Figure 1. Apparatus for flow visualization and relative permeability measurement of rough-walled rock fracture. The fracture specimen is indicated by the shaded area; its approximate size is 3" x 3".

Flow experiments have been carried out with transparent fracture replicas, allowing visualization, and with actual rock fractures where flow behavior is not visible, but is characterized by rate and pressure data. Among our significant qualitative findings is the demonstration and explanation of persistent instabilities in two-phase flow. As an example, Fig. 2 shows pressure data for flow in a transparent replica of a fracture found in a granite core from Stripa, Sweden. Persistent cycling of gas inlet and outlet pressures is observed for conditions of constant applied gas and liquid flow rates. Visual observation revealed that gas flow occured through a single path that was periodically blocked and unblocked by water at a particular pore throat. To discuss the flow mechanism suppose that, at a certain time, the smallest ("critical") pore throat along a potential gas flow path from inlet to outlet is blocked by water.

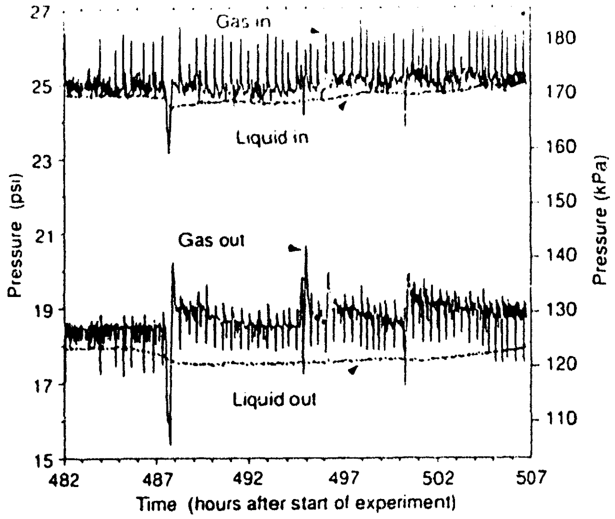

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Figure 2. Observed pressure cycling for constant-rate gas injection in a replica of a fracture in granite from Stripa, Sweden.

Constant-rate gas injection will cause gas pressures to increase upstream from the throat, until gas pressure P_g exceeds liquid pressure P_l in the throat by more than the capillary entry pressure, $P_{ent} = 2\sigma/r_c$, where σ is the surface tension of water, and r_c is the radius of the critical throat. When $P_g > P_l + P_{ent}$, gas will invade the throat and will migrate downstream to establish a continuous gas flow path towards the outlet. Gas pressures upstream from the throat will then drop substantially, whereupon the critical throat will be re-invaded by liquid, returning to a blocked condition and repeating the cycle. We observed such pressure cycling behavior for periods of many days. It is of interest to note that pressure cycling in two-phase flow has also been observed during well tests at NAGRA's field site in Wellenberg/Switzerland,[6] and in numerical simulation experiments on networks of capillary tubes.[7]

By injecting gas at constant pressure rather than at constant rate we have been able to achieve very stable (non-cycling) two-phase flows, and to obtain measurements of gas and liquid relative permeabilities as function of applied capillary pressure. Fig. 3 shows measured relative permeabilities for a natural granite fracture, and for a replica of a tuff fracture, plotted in the form of liquid vs. gas relative permeability. It is seen that the sum of liquid and gas relative permeabilities is much less than 1 for most of the data, indicating that the interference between the two phases is very strong. This result runs counter to the "conventional" view of fracture relative permeabilities, which held that the sum of liquid and gas relative permeabilities should be approximately equal to 1 at all saturations.[8] However, the observed strong phase interference is in qualitative agreement with recent theoretical models that view "small" fractures as two-dimensional heterogeneous porous media.[9]

The work on two-phase fracture flow visualization and measurement is ongoing. Current experimental efforts use fracture specimens from Yucca Mountain tuffs, and aim at flows on a larger spatial scale.

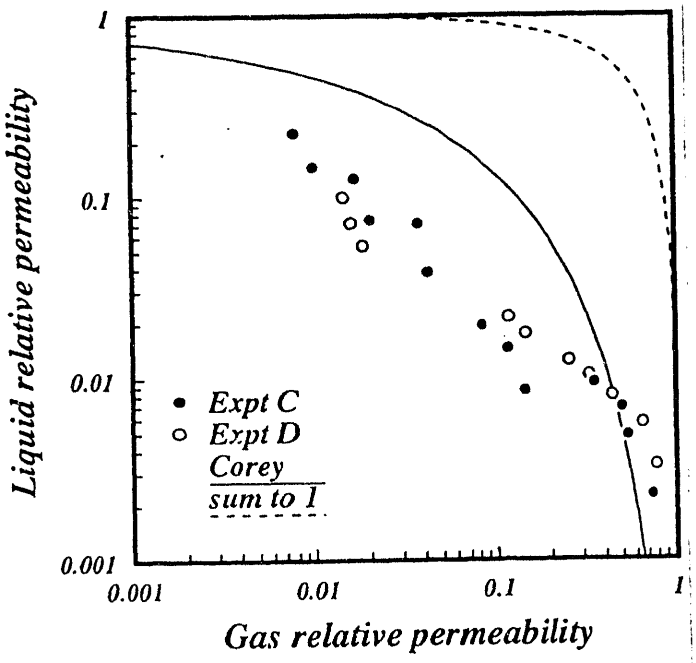

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Figure 3. Measured gas and liquid relative permeabilities. C: natural fracture from Stripa, D: replica of fracture from Dixie Valley.

III. MODELING OF TWO-PHASE FLOW IN HETEROGENEOUS MEDIA

Water infiltration in the unsaturated zone, as at Yucca Mountain, and gas release from beneath the water table, are gravitationally unstable processes. They can give rise to highly channelized flow along preferential pathways, especially in heterogeneous and fractured media. Such flows could permit rapid migration of contaminants.

We are performing theoretical studies and numerical experiments to better understand the nature of gravity-driven immiscible displacement in heterogeneous fractured media. Ongoing investigations address flow processes on different spatial scales, from the development and migration of individual liquid and gas "fingers" to global average behavior of liquid infiltration plumes.

When aqueous or nonaqueous fluids infiltrate into the vadose zone, liquid saturation near the injection point will increase. Liquid saturation may rise all the way to 100 %, establishing single-phase conditions with pressure buildup and consequent lateral flow. If the permeability of the medium is sufficiently high, or liquid fluxes sufficiently low, the medium will remain in two-phase conditions. Then under isothermal conditions no pressure buildup will occur, and liquid flow will be affected only by gravity and capillary forces. In media with large pores, such as coarse-grained soils, or "large" fractures in hard rocks, capillary effects tend to be weak, and water flow will be dominated by gravity effects. (Water flowing downward in fractures may also be subject to capillary-driven imbibition into the low-permeability rock matrix.) For gravity-dominated flow water will move primarily downward, but "straight" downward flow is only possible when appropriate permeability is available in the vertical direction. Water flowing downward in coarse soils, or in large (sub)vertical fractures, may encounter low-permeability obstacles, such as silt or clay lenses in soils, or asperity contacts between fracture walls. Water will pond atop the obstacles and be diverted sideways, until other predominantly vertical pathways are reached (Fig. 4).

The conventional treatment of two-phase flow includes gravity, pressure, and capillary effects. It employs a multiphase version of Darcy's law with mass fluxes $\mathbf{F}_\beta$ (β = liquid, gas) given by

$$F_\beta = -\mathbf{k}\frac{k_{r\beta}}{\mu_\beta}\rho_\beta\left(\nabla P_\beta - \rho_\beta \mathbf{g}\right) \qquad (1).$$

Here, **k** is the permeability tensor, $k_{r\beta}$ is relative density, P_β is pressure in phase β, and g is acceleration of gravity. Horizontal flow diversion from media heterogeneities can be represented only if such heterogeneity is modeled in full explicit detail. In practical applications heterogeneities occur on many different scales (impermeable lenses, individual fractures, fracture networks, capillary barriers, lithologic units, etc.). Detailed characterization and modeling would be prohibitively complex, and simplified approaches are needed to capture the essential effects.

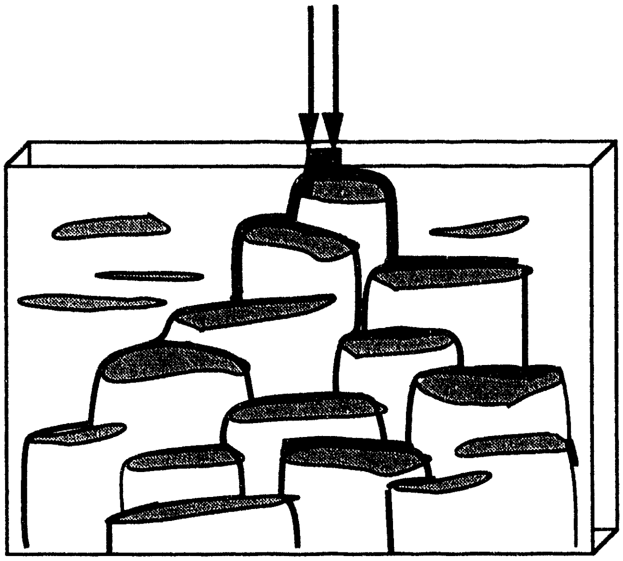

Figure 4. Schematic of liquid infiltration in an unsaturated heterogeneous medium. Regions of low permeability (shaded) divert flux sideways and cause a lateral spreading of the infiltration plume.

We have developed an extension of the conventional two-phase flow theory that proposes to approximate the dispersive spreading of liquid infiltration plumes as a Fickian diffusion process. In analogy to solute dispersion in miscible flow,[10] we add a dispersive flux term to Eq. (1) which is taken to be proportional to the gradient of liquid saturation, S_l.

$$\mathbf{F}_{l,\mathrm{dis}} = -\rho_l \, \phi \, \mathbf{D}_{\mathrm{dis}} \nabla S_l \qquad (2).$$

ϕ is the medium porosity, and the dispersion tensor $\mathbf{D}_{\mathrm{dis}}$ is written as[11]

$$\mathbf{D}_{\mathrm{dis}} = v\left(\alpha_T\left[\mathbf{e}_x \mathbf{e}_x + \mathbf{e}_y \mathbf{e}_y\right] + \alpha_L \mathbf{e}_z \mathbf{e}_z\right) \qquad (3).$$

In Eq. (3) we have introduced unit vectors **e** in the x, y, and z-directions, and have written transverse and longitudinal dispersion coefficients in the usual way as products of transverse (horizontal) and longitudinal (vertical) dispersivities α_T, α_L, and an advective velocity v for the propagation of saturation disturbances.[12]

To examine the validity of the Fickian dispersion hypothesis we have performed numerical simulation experiments in media with fully-resolved small-scale heterogeneity. The calculations were done with our multiphase flow code TOUGH2,[3] enhanced with a set of preconditioned conjugate gradient routines for efficient solution of multidimensional flow problems with 10,000 or more grid blocks (Moridis, private communication, 1993). The flow experiments involve placing a localized plume of liquid into an unsaturated heterogeneous medium, such as shown in Fig. 5, and then allowing the plume to migrate under the combined action of pressure, capillary, and gravity forces (Eq. 1). Small-scale medium heterogeneity is resolved in detail, and no explicit allowance for phase dispersion as in Eq. (3) is made.

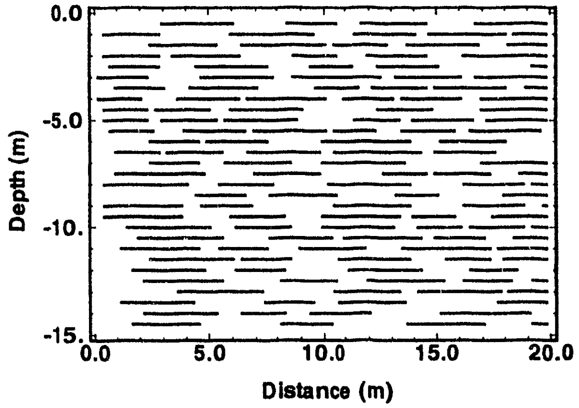

Figure 5. Two-dimensional vertical section of a heterogeneous medium with a random distribution of impermeable obstacles. Length of obstacles is uniformly distributed in the range of 2-4 m.

Many numerical experiments were carried out for media with different parameters and style of heterogeneity, both regular and stochastic. As an example, Fig. 6 shows simulated liquid infiltration plumes after 2×10^5 seconds in the medium of Fig. 5 (permeability of 10^{-11} m^2). When capillary pressures are neglected (or unimportant, as in coarse high-permeable media or large fractures), flow proceeds in the form of narrow fingers.[13] Capillary suction pressures will tend to dampen out the narrower fingers. By calculating spatial moments for the simulated infiltration plumes,[14, 15, 16] we have determined transverse dispersivities, see Fig. 7.

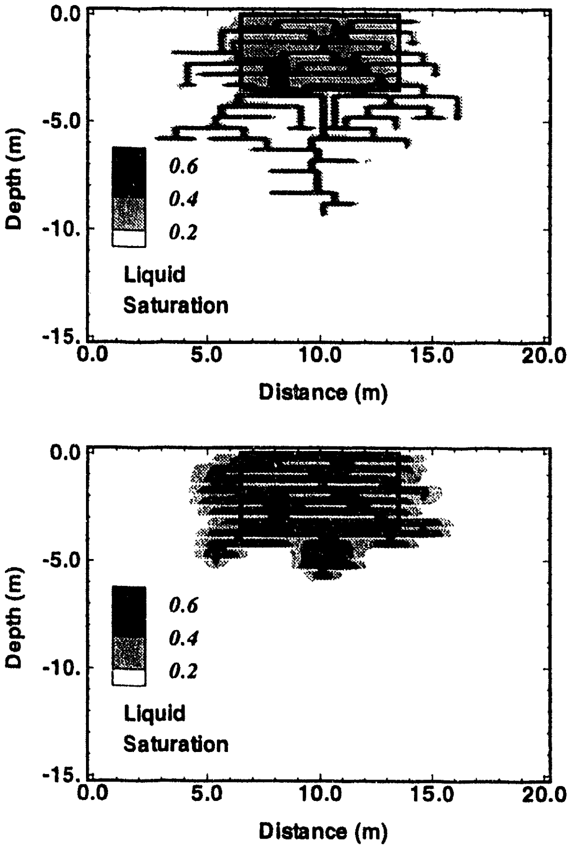

Figure 6. Simulated infiltration plumes in the medium of Fig. 5 after $2x10^5$ seconds. The plume on top is for a case without capillary pressure, the plume on the bottom includes capillary pressure effects. Initially the plume has a uniform water saturation of $S_l = 0.99$ and occupies the region indicated by the black rectangle at the top of the figures.

The most important result is that transverse dispersivities stabilize, after a period of transient changes at early times, at nearly constant values. This stabilization occurs regardless of the strength of capillary pressure, and indicates that transverse plume spreading from the intrinsic heterogeneities of the medium indeed gives rise to a Fickian diffusion process. Many additional simulations have been performed that suggest that broad classes of heterogeneous media disperse infiltrating liquid plumes transversally in a Fickian manner. However, it should be emphasized that Fickian-type dispersive behavior from medium heterogeneities is by no means inevitable or universal. In fact, for certain heterogeneity conditions and spatial scales infiltration plumes may show "anti-dispersive" behavior, becoming more narrowly focussed with depth.[17] Flow behavior depends on the nature of the heterogeneities and the strength of capillary forces.[18]

The above discussion dealt with phase dispersion effects from heterogeneity for descending liquid infiltration plumes. Similar kinds of effects are expected to develop when gas plumes released beneath the water table rise in the saturated zone. Further numerical and physical experiments, and field observations, are needed to determine the range of heterogeneity conditions under which the Fickian dispersion model is applicable.

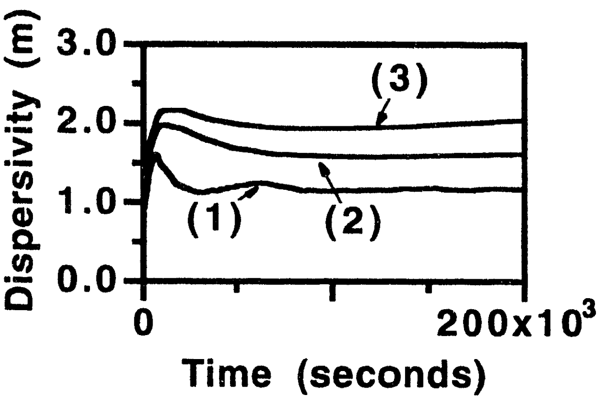

Figure 7. Transverse dispersivities for the plumes of Fig. 6 for cases with increasingly strong capillary pressures: (1) no P_{cap}, (2) moderate P_{cap}, (3) strong P_{cap}.

IV. DESIGN AND ANALYSIS OF FIELD EXPERIMENTS

Regular ventilation during repository operation will lead to water evaporation and partial desaturation of the rock immediately surrounding the excavations. By monitoring air humidity in ventilated tunnel sections it is possible to measure liquid flow rates to the drifts. This allows estimation of hydraulic properties of the formation on a scale relevant to flow in the near-field of a repository. Combined ventilation and brine injection tests are being conducted at the Grimsel Rock

Laboratory, Switzerland. The objective of the experiments is to study the transport of liquid and gas in the vicinity of a ventilated drift in order to evaluate the impact of the drying process on the characterization of the rock matrix.

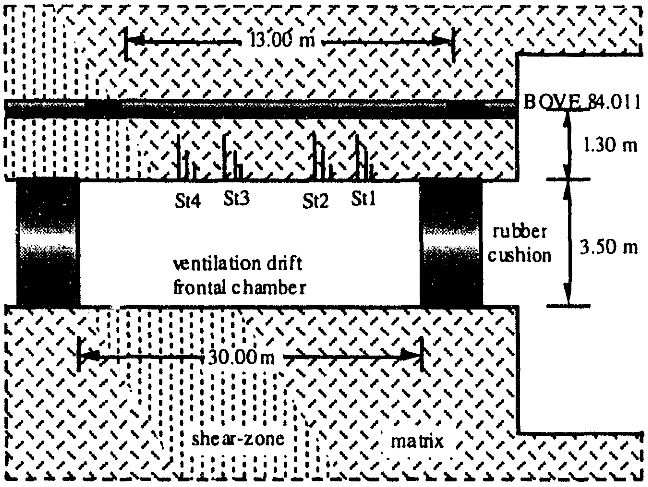

Figure 8. Schematic plan view of ventilation drift, borehole, and test equipment.

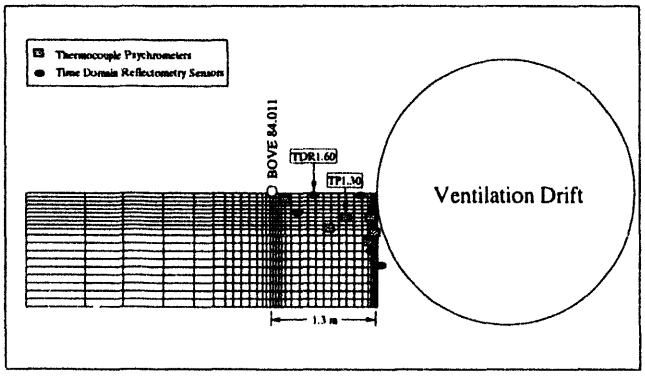

Figure 9. Model domain and computational grid; vertical cross section through St1.

The proposed test sequence includes a desaturation - resaturation cycle. In addition, brine and fresh water will be injected from a borehole as trace electrolytes in order to better track the propagation of the individual phases. The geometrical layout of the drift and the boreholes as well as the major structure features is depicted in Figure 8. Borehole BOVE 84.011, which is parallel to the drift, has been selected for brine and water injection. Water potential, liquid saturation, and salt concentration are observed at four locations along the drift (St1 - St4), each station being equipped with six thermocouple psychrometers (TP) and two tensiometers at different depths and up to six time domain reflectometry sensors (TDR) for measuring the dielectric constant and the bulk electric conductivity. A simplified two-dimensional rectangular grid (Figure 9) is employed for numerical analysis by means of the ITOUGH2 code,[19] an inverse version of TOUGH2. ITOUGH2 provides automatic model calibration to data from a previous ventilation experiment. Measurements of negative water potentials at a depth of 2, 5, 10, 20, 40, and 80 cm over a period of eighty days are used to calibrate the model by estimating absolute permeability as well as parameters of van Genuchten's relative permeability and capillary pressure functions. Figure 10 shows the comparison between measured and calculated system response.

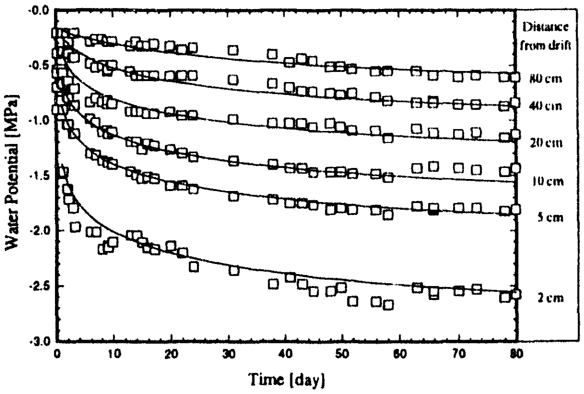

Figure 10. Fit between computed (lines) and measured (symbols) water potentials

The permeability of the granodiorite matrix is a low 7×10^{-19} m^2, so that capillary pressures are very strong even for low gas saturations. Heterogeneity may induce partial drying of the formation and consequently preferential flow of brine and water. Heterogeneity effects are explored by introducing a linear feature, perpendicular to the drift, with higher permeability and lower air entry pressure compared to that of the surrounding rock matrix. The calibrated model is then used in "forward mode" to predict saturation, capillary pressure, and tracer response. The following test sequence has been modeled:

(1) The granodiorite at the Grimsel Test Site is believed to be fully liquid saturated prior to ventilation.

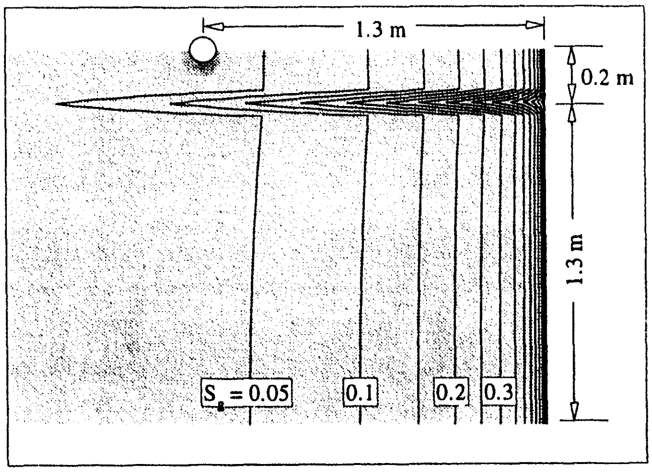

Figure 11. Gas saturation after 6 months of ventilation.

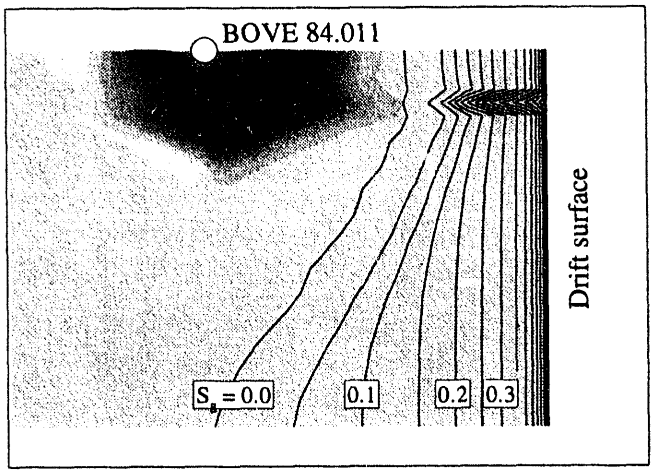

Figure 12. Brine content (shaded surface) and gas saturation (contours) after 3 days of brine and 3 days of fresh water injection

(2) The relative humidity in the drift is reduced by ventilation to about 68 %, leading to an equivalent capillary suction of -50 MPa at the drift wall. Evaporation occurs at the surface, and the rock matrix is allowed to desaturate over a period of six months. Due to the weaker capillary pressure in the high-conductive feature, liquid is imbibed into the adjacent rock matrix leading to higher gas saturations in the fracture zone (see Figure 11).

(3) Subsequently, a 0.05 molar NaCl solution is injected in borehole BOVE 84.011 at a constant rate for three days, followed by a three-day period of fresh water injection at the same rate. Injecting liquid leads to reduced gas saturation in the vicinity of the borehole, and brine and water flow preferentially along the high conductive zone as indicated in Figure 12.

(4) Injection and ventilation stops, and the propagation of the gas-brine and brine-water front is observed during resaturation of the formation.

A detailed discussion of the simulated test sequence, including sensitivity studies and an analysis of prediction errors, can be found in a recent LBL-report.[20] The results of these design calculations using the TOUGH2 code show that injection of brine may significantly influence the unsaturated flow behavior by changing the pressure and saturation distribution around the borehole. Transport velocity is predicted to be very slow, requiring several months for the brine to reach the drift wall. However, the presence of preferential flow paths may reduce travel time and alter brine content and saturation distribution.

Numerical simulations as shown herein help improve the understanding of basic two-phase flow processes. A more comprehensive discussion of inverse modeling for calibration and site characterization is provided in a companion paper.[21]

V. RESULTS AND CONCLUSIONS

Two-phase flow processes as water descends in the unsaturated zone, or gas rises in the saturated zone, are expected to play an important role in subsurface disposal of nuclear wastes. These processes are characterized by gravitational instability of denser over less dense fluid, and show a strong sensitivity to permeability and capillary heterogeneity of the (generally fractured-porous) media. There is a potential for highly channelized flows along preferential pathways, with rapid migration of contaminants.

By combining laboratory and field studies with theoretical analysis and modeling, we are attempting to identify important flow phenomena, and to characterize them conceptually and quantitatively. Our two-phase laboratory flow visualization experiments in transparent replicas of rough-walled natural rock fractures have revealed a tendency towards cyclic instability in two-phase flows, with continual blocking and unblocking of critical flow paths. These phenomena have been explained in terms of an interplay between gas-liquid capillary pressures and pressure drops in viscous flow. Gas and liquid relative permeabilities were measured for transparent replicas as well as for actual rock fractures. Results obtained so far indicate "strong phase interference," i.e., relative permeabilities of both phases are small at intermediate saturations, even smaller than for typical porous media. These results contradict the conventional view of fracture relative permeabilities, but they are in qualitative agreement with theoretical predictions from conceptualizations of fractures as two-dimensional heterogeneous porous media.

Numerical simulation experiments have demonstrated that ever-present medium heterogeneities will cause a lateral (transverse) spreading of descending liquid infiltration plumes. Quantitative analysis showed that for media with random heterogeneity the plume spreading can be described as a Fickian diffusion process. This finding supports a recently proposed extension to two-phase immiscible flow theory, in which a phase-dispersive flux term is added to the usual pressure, capillary, and gravity-driven flows.

One of the most difficult aspects of two-phase flow is characterization of a site from field tests and observations. Inverse modeling techniques have been developed that can utilize different kinds of experimental data to perform an automatic calibration of a proposed conceptual model. Quantitative information for judging model acceptability is also provided. These techniques were applied to long-term (months) ventilation tests to develop a calibrated quantitative model for two-phase flow processes in a section of the Grimsel Rock Laboratory, Switzerland. The model was then used in forward (predictive) mode to design further gas and tracer injection experiments.

In closing, it should be emphasized that two-phase flow systems are capable of a great complexity and diversity of behavior on different space and time scales. This paper has only been able to address a very limited set of issues. Continued laboratory, field, and theoretical efforts are needed to learn more about the dependence of two-phase flows on the properties of the permeable medium, the initial and boundary conditions, and the nature of external perturbations (natural or man-made). Only through continued vigorous research will it be possible to develop a scientific basis for site characterization, site suitability evaluation, and engineering design decisions.

ACKNOWLEDGEMENTS

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