

**EVALUATION OF THE POST-EMPLACEMENT ENVIRONMENT OF HIGH
LEVEL RADIOACTIVE WASTE PACKAGES AT
YUCCA MOUNTAIN, NEVADA**

William Glassley
Lawrence Livermore National Laboratory
Livermore, California 94550

ABSTRACT

Evaluation of the post-emplacement environment around high level radioactive waste containers is required by federal regulations. The information derived from this evaluation will be used to determine the service performance of the waste containers, the chemical and hydrological conditions that may influence radionuclide release and transport if containers are breached, and retrievability of the waste containers prior to closure of the repository. Laboratory studies, numerical simulations, and field experiments and tests are used to provide data necessary for this evaluation. Results obtained to date demonstrate that the post-emplacement environment in the welded tuff at Yucca Mountain, Nevada maintains relatively benign chemical features (i.e., near neutral pH, low concentrations of dissolved species) for most scenarios. The hydrological environment appears to be one of low flow volume and rates for the expected condition of an unsaturated medium. Emplacement borehole stability will be a function of fracture density and orientation, which may be influenced by microcrack development. Field studies and numerical simulations are in progress that will extend the results of laboratory studies to long time periods. The extent to which chemical, hydrological and mechanical processes can be adequately coupled through numerical simulations remains a matter of concern.

The objective of the waste package environment studies is to establish and characterize the environmental processes affecting the near-field repository host rock and fluid after waste package emplacement. These processes, which reflect the perturbation induced in the environment by engineering effects and by the waste package decay heat and radiation, will influence chemical, mineralogical, hydrological, and mechanical features of the environment. To assure that waste package design considerations reflect the characteristics of this evolving environment it is necessary to determine the range of conditions that may develop in the pre- and post-emplacement waste package setting.

Three broad categories of activities provide the information necessary to characterize the pre- and post-emplacement waste package environment. These categories respectively establish the water quality that may contact waste packages, the quantity of water that may contact waste packages, and thermo-mechanical properties of tuff in the vicinity of waste packages (Fig. 1). These activities will provide information that will be used to evaluate the performance of the waste packages, and will be used in developing tests and selection criteria for evaluating and designing the metal barriers.

**SUMMARY OF ENVIRONMENTAL
CHARACTERISTICS**

The proposed environment in which high level radioactive waste would be emplaced is a devitrified, welded, rhyolitic tuff that consists of primary minerals, such as sanidine, plagioclase, quartz, biotite, iron-titanium oxides, allanite and zircon, that formed at temperatures in excess of 600°C in a magma chamber prior to eruption of the tuff, and secondary minerals, such as cristobalite, quartz, alkali feldspars, and smectite clays that formed during cooling and later alteration of the tuff at temperatures less than 500°C. These minerals form a physical framework that contains pores partially filled with fluid, as indicated by the fact that the rock in the proposed repository horizon has a mean matrix porosity of 11-14% and a mean water saturation of 65% (1,8). Modeling of the thermal behavior of the waste package near-field environment demonstrates that there will be a rapid increase in temperature immediately after package emplacement (2). The maximum temperature attained will depend on the nature of the waste emplaced and the repository design, but is expected to be approximately 230°C. The temperature is then expected to slowly decrease over hundreds of years. This thermal behavior will result in development of a dehydration zone surrounding the waste

package as water vaporizes and migrates away from the waste package.

This environment will modify the thermal and radiation fields experienced by the host rock, and may lead to complex chemical reactions, mechanical responses, and fluid flow regimes. To accomplish thorough characterization of the environment, it is necessary to evaluate the effects of temperature, nature of the fluid phase (liquid and/or steam in the presence of air), dissolution and precipitation kinetics, radiation dose and dose rate, rock surface area to fluid volume ratio, pore water chemistry, fluid flow pathways, flow rates and volumes, and microcrack and fracture behavior in the perturbed environment. The range of values appropriate for these parameters in the post-emplacement environment, and the effects these parameters have on the evolution of the environment, are the focus of activities in this technical area.

NEAR-FIELD MINERALOGY AND CHEMISTRY

Rock-Water Interaction

The rock-water system adjacent to the waste package will experience a thermodynamic drive to attain equilibrium with the new thermal and chemical environment. The thermodynamic drive will be expressed as chemical reactions

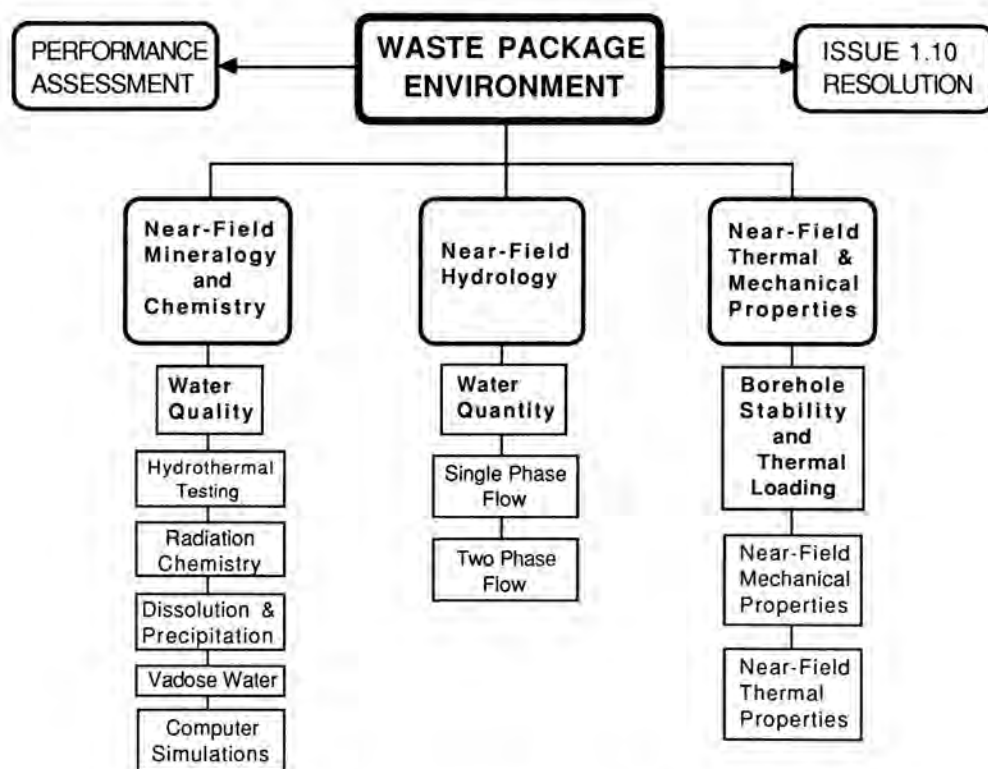


Fig. 1. Waste Package Environment Model and Activity Hierarchy.

among the mineral and fluid phases in the environment. Depending upon the extent of reaction, detectable dissolution of host rock primary and secondary minerals may occur, solid mineral phases may form, and mineral and fluid compositions may change. The extent to which these reactions proceed will depend upon the kinetics of dissolution, precipitation, and diffusion processes.

A series of long-term tests (3) have been conducted over a range of temperatures (90 to 250°C) using drill core material. These tests demonstrate that the solid phase reaction products that form during interaction tuff with reference ground water at elevated temperatures are zeolites, clays, and carbonates. Steady state fluid compositions are approached, thus providing an indication of the composition of the fluid that would be expected to exist in the vicinity of waste packages. These fluids are usually saturated in cristobalite, and have low concentrations of other cations (Fig. 2). Conducting these tests over a range of temperatures establishes the stability fields of reaction product minerals and solid solution effects. Furthermore, because water confined in the rock pores may exist to temperatures as high as 140°C if venting is restricted (4) experiments conducted at high temperatures can help define the nature of fluids and reaction products that can develop in these small pores.

Other effects that are also evaluated in these and other experiments include the ratio of rock surface area to fluid volume, the effect of reaction progress, and the phases in

the host rock that dissolve or change in composition during the reaction process.

Radiation Effects

Studies of rock-water interaction in the presence of a gamma radiation field are being conducted to establish the identity and quantity of solid, gaseous, and dissolved reaction products. The purpose of these tests and experiments is to determine the effect radiation will have on the chemical and mineralogical characteristics of the waste package environment.

Of particular concern for container and waste form performance is whether a bicarbonate-buffered aqueous solution (such as J-13 water) in contact with air will form formate or oxalate. These tests and experiments will also establish whether the nitrite-nitrate ratio changes if MnO_2 is present, the extent to which the repository horizon tuff mineralogy will buffer the fluid pH if nitric acid is produced through radiolytic processes, the mechanism of nitric acid formation, and the reactions that occur when NO dissolves in the liquid phase. All of these processes affect the chemical evolution of the fluid phase and directly contribute to establishing the properties of the fluid in the waste package environment.

Pore Water Composition

Circumstantial evidence exists that the pore water in the repository horizon is similar in composition to water obtained from well J-13, which is the YMP reference

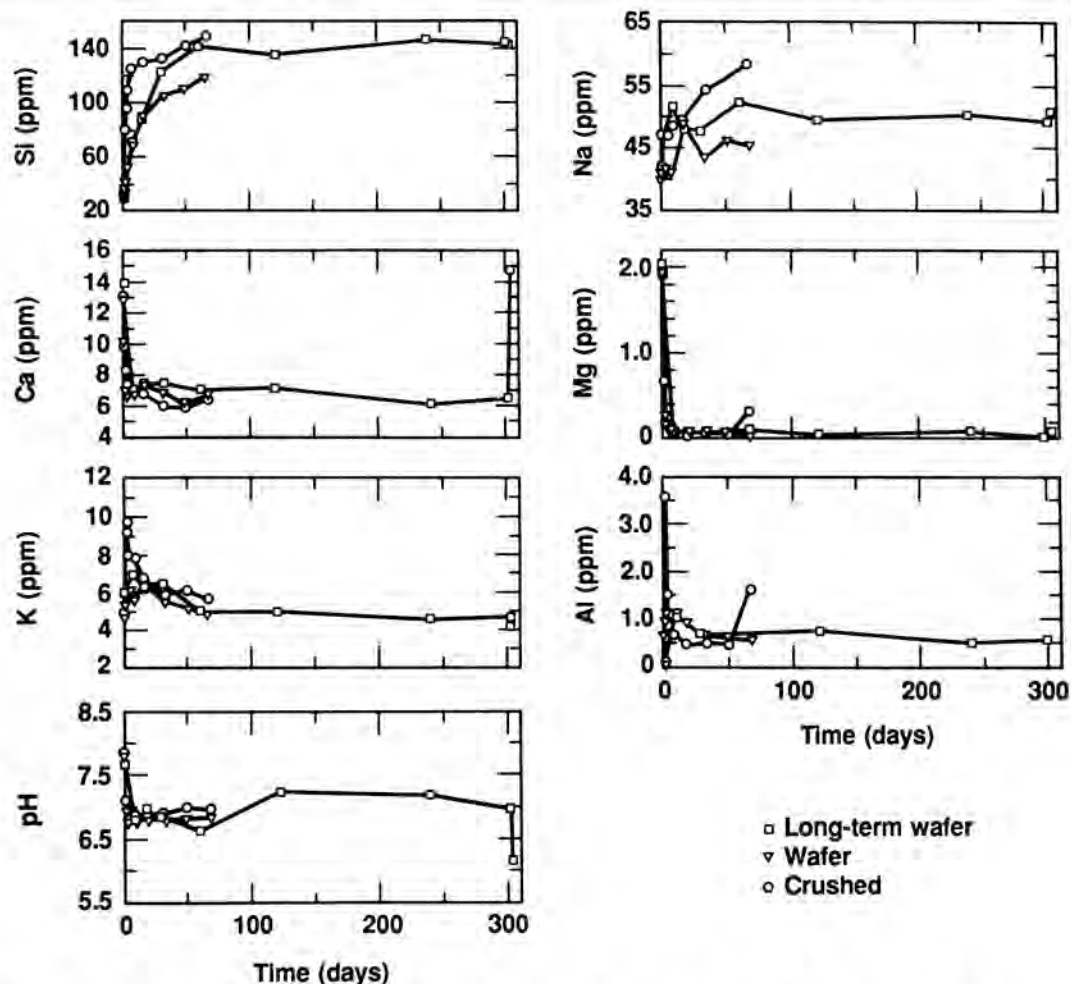


Fig. 2. Chemical Composition of Fluid in Contact with Tuff Samples in Gold-Bag, Rocking-Autoclave Experiments Conducted at 150°C (3). Changes at Day 300 are Analyses of Quench Samples When Experiment was Terminated.

groundwater (Table I). This evidence consists of correlations between rock and fluid compositions at Rainier Mesa, which is geologically similar to Yucca Mountain. However, experimental work must be done to confirm that the vadose water at the repository horizon is, in fact, similar in composition to the reference ground water. Fluid extraction techniques that have been employed in the past include high pressure compression of core samples, application of high gas pressure to core samples, centrifugation, and pore water displacement (5,6).

Numerical Simulations

The geochemical behavior of the waste package environment over long time periods (tens to thousands of years) will vary as the radiation and thermal fields change through time. Laboratory study is incapable of examining the coupled effects of all possible values and combinations of parameters significant for the system under consideration. In addition, laboratory studies cannot be directly used to extend the behavior of natural systems studied in the laboratory over periods of months to periods of many years. However, the use of numerical geochemical simulations, in conjunction with judiciously selected tests, can provide the

capability to examine effects and processes in natural systems for time periods and chemical conditions not duplicated by experimental studies. To accomplish the range of geochemical simulations necessary to complete the modeling effort, activities have been designed that will 1) establish the most effective and efficient approach to numerical simulations of the tuff-water system, 2) conduct validation simulations to establish the validity of the computational methodology and database values to be applied to the system under examination, and 3) model long-term rock-water interaction.

Initial numerical simulations, using EQ3/6, of the evolution of rock-water systems have been completed on several hydrothermal studies (7). The efficacy of the modeling approach has been established by duplicating many of the observed changes in fluid composition and mineral development noted in the experiments. In addition, certain limitations in the thermodynamic database relating to precipitation kinetics and standard state data were

TABLE I
Compositions of Interstitial, Fracture, and Tunnel Water, Compared to Well J-10

	<u>Interstitial (16)</u>	<u>Fracture (16)</u>	<u>Tunnel Water (17)</u>	<u>Well J-13 (18)</u>
Na	1.73	1.53	2.30	1.96
K	0.18	0.12	0.11	0.14
Ca	0.27	0.21	0.08	0.29
Mg	0.10	0.06	0.01	0.07
HCO ₃ ⁻	1.14	1.61	2.25	2.34
SO ₄ ⁼	0.43	0.15	0.10	0.19
Cl ⁻	0.75	0.24	0.18	0.18
SiO ₂	0.97	0.88	0.73	1.07
pH	7.8	7.5	7.0	6.0

(All values except pH are in mmoles/liter.)

(Numbers in parentheses refer to reference list.)

identified. Upon incorporation of new thermodynamic data for clays and zeolites, more accurate modeling of reaction progress is expected.

Currently under evaluation are analogous natural systems that may be used to validate the calculational approach. The natural systems selected for the validation activity will encompass a broad range of compositions and conditions. Use of a broad spectrum of compositions and conditions in the validation activity assures that natural variation in the repository system will be accounted for, that the thermodynamic and kinetic attributes of the package environment are adequately described, and that limitations to the results of numerical simulations are rigorously defined. In addition, these simulations will include natural systems that have been active for thousands or millions of years, thus providing constraints on the kinetics effects that may be influential in the waste package environment.

These modeling activities will also provide input to waste package performance assessment by establishing the composition of fluid in the waste package environment for the anticipated and unanticipated range of repository conditions over the time interval defined by regulatory requirements. The code and database applications will also be used to determine the sensitivity of the geochemical environment to perturbations in individual compositional parameters within the range for which validation was accomplished. Results of this analysis will define limits of variation in the

geochemical system that will constrain the input to waste package performance assessment.

NEAR-FIELD HYDROLOGY

The hydrological properties of the tuff are a reflection of the degree of welding, extent of secondary mineral growth, magnitude of fracturing, and degree of saturation. These properties have resulted in a rock with a mean matrix porosity of 11-14% (1,8) and a fracture density of between 8 and 40 fractures per cubic meter (9). The structural location of this horizon 700 to 1400 feet above the local water table (10), and the local meteorological conditions have resulted in a rock that is approximately 65% saturated (1). Evidence suggests that the net water flux in this unsaturated environment is 1.0 to 2.0 mm/yr upward, although a downward flux of 1.0×10^{-7} to 0.5/mm/yr may occur as a result of matrix flow (1,11). The matric potential of the tuff is approximately -112 kPa, which results in negligible fracture flow (12).

Experiments completed to date demonstrate that the absolute permeability of the fractures and intact tuff differ by at least three orders of magnitude (13,14). This contrast in permeabilities (and in other hydrological properties) leads to a complex hydrological environment in which flow via matrix or fracture porosity is a sensitive function of degree of saturation, fracture aperture and fracture roughness. In addition, laboratory studies have demonstrated the

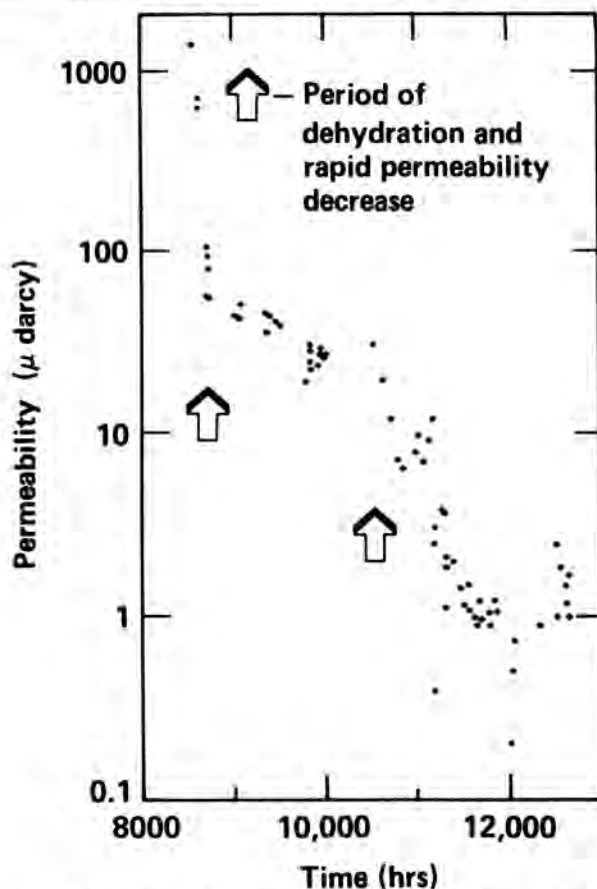


Fig. 3. Change in Permeability With Time in a Fractured Sample of Tuff. The Permeability Changes, Which are Indicated by the Arrows, Occurred During Dehydration Episodes Imposed on the Core. The Core Sample was Contained in a Pressure Vessel, and was 8.23 cm in Diameter (14).

possibility that fractures may seal during dehydration/rehydration cycles (Fig. 3).

Numerical simulations of the behavior of fluid during heating and cooling of the tuff in the vicinity of the waste packages is being approached via continuum and discrete particle methodologies. The continuum approach uses an integrated finite difference computational scheme that tracks the evolution through time of the thermal field and fluid flow regimes. The particle tracking approach (Fig. 4) derives the motion of individual fluid packets in a medium of known hydrological properties. The extent to which these approaches allow simulation of fluid flow in the thermally perturbed environment is currently under evaluation.

MECHANICAL PROPERTIES

Keyblock analysis (15) has indicated that emplacement borehole stability will be maintained if the fracture geometry and frequency is sufficient to develop blocks of dimensions greater than the borehole diameter. However, thermal perturbation of the environment may result in the development of microcracks that may effect spallation or fragmentation of borehole walls. Abrasion of fracture surfaces may also occur during thermal expansion and contraction, resulting in changes in the surface characteristics of the fractures. Field and laboratory studies are currently under-

way to evaluate the consequences for emplacement borehole stability of these processes.

FIELD-ORIENTED EFFORTS

The extent to which laboratory studies apply to natural systems can only be established by conducting a field testing program. We have undertaken an extensive simulation effort in which large heaters are emplaced in boreholes in tuff and the resulting temperature, fluid movement, and rock behavior are remotely monitored. A variety of sensing techniques, including HFEM tomography, and neutron and gamma probes are being evaluated in these long duration tests at G-tunnel, in preparation for simulations of waste package emplacement at the Exploratory Shaft Test Facility. These tests will provide a direct evaluation of the extent to which laboratory studies can be extrapolated to the complex natural setting of the proposed repository, and also provide an opportunity to fully exercise the predictive capabilities of the computer codes used to model the long-term behavior of the repository environment.

CHALLENGES

The greatest challenge faced by this effort is developing the capability to couple fluid flow with chemical reaction processes. Although static reaction path modeling has been developed and applied to the tuff/water system, accurately simulating fluid flow in a system of variable permeability

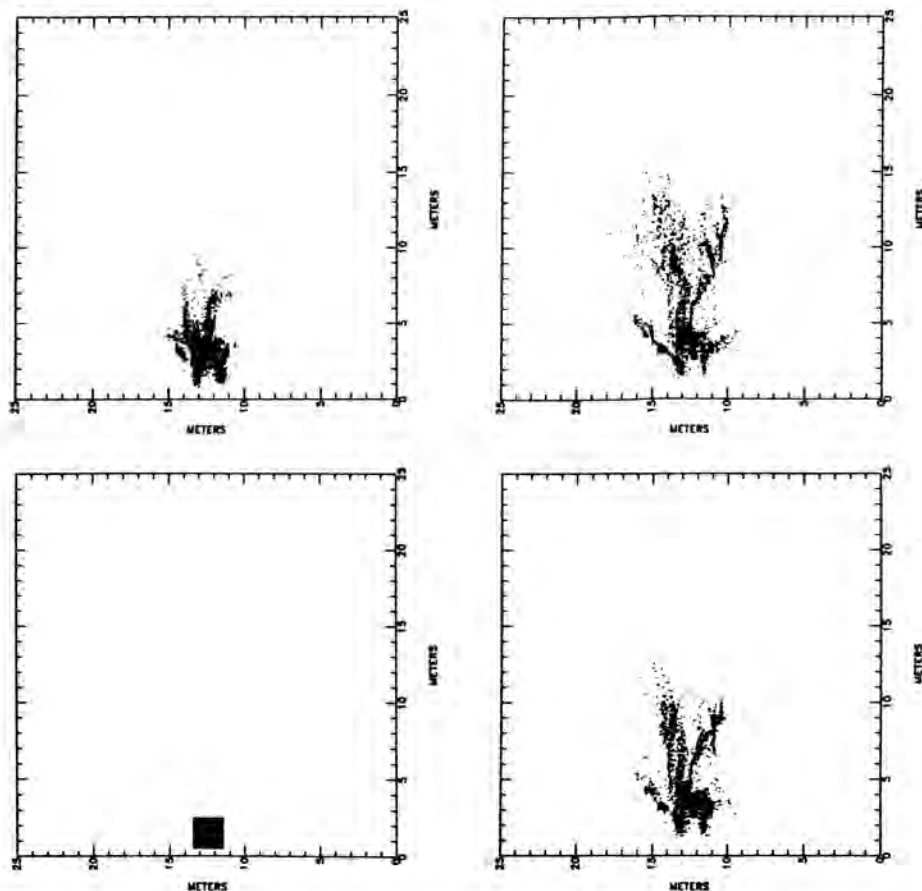


Fig. 4. Two-Dimensional Projection of a Simulated Release of Particles into a Flow Field Passing Through a Saturated, Porous Medium. Time Progresses From Upper Left to Lower Right. Simulation Performed by Andrew Thompson, Lawrence Livermore National Laboratory.

under unsaturated conditions has yet to be adequately accomplished. Nor has it been possible to simulate accurately reactive processes in an unsaturated environment in which fluid flow occurs. Coupling the hydrological processes with the chemical processes thus remains a necessary but elusive goal, which may take many years to attain.

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