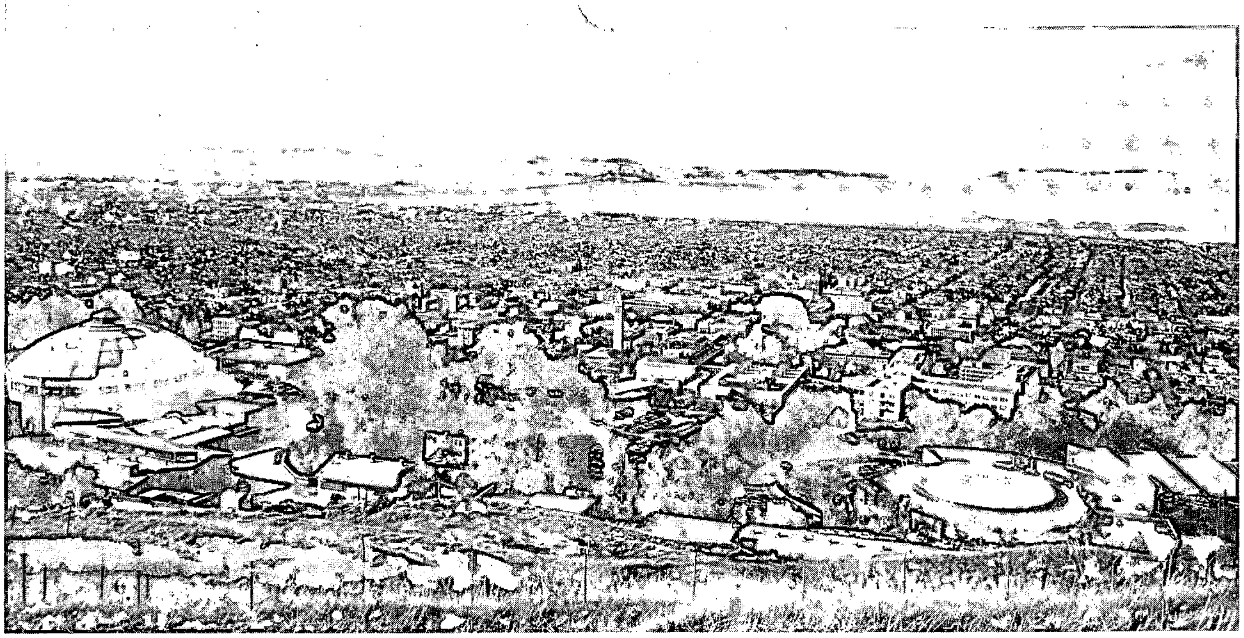
Lawrence Berkeley Laboratory
UNIVERSITY OF CALIFORNIA

EARTH SCIENCES DIVISION

Report of the Second Meeting of the Consultants on Coupled Processes Associated with Geological Disposal of Nuclear Waste, Lawrence Berkeley Laboratory, Berkeley, CA, January 15–16, 1985

C.-F. Tsang and D.C. Mangold

September 1985



Prepared for the U.S. Department of Energy under Contract Number DE-AC03-76SF00098.

LBL-19456

REPORT OF THE SECOND MEETING OF THE CONSULTANTS ON COUPLED PROCESSES ASSOCIATED WITH GEOLOGICAL DISPOSAL OF NUCLEAR WASTE

Lawrence Berkeley Laboratory
January 15-16, 1985

Consultants

Bruce R. Clark
Neville G. W. Cook
Ghislain de Marsily
Robert O. Fournier
Kalman Kovari
Donald Langmuir
Chin-Fu Tsang
Paul A. Witherspoon, Chairman

Rapporteurs:
Chin-Fu Tsang
Donald C. Mangold

September 1985

Earth Sciences Division
Lawrence Berkeley Laboratory
University of California
Berkeley, California 94720, USA

CONSULTANTS ON COUPLED PROCESSES ASSOCIATED WITH GEOLOGICAL DISPOSAL OF NUCLEAR WASTE

Dr. Bruce R. Clark
Leighton and Associates, Inc.
1151 Duryea Avenue
Irvine, California 92714
(714) 250-1421
or (213) 691-2125

Dr. Neville G. W. Cook
Department of Materials Science
and Mineral Engineering
University of California
Berkeley, California 94720
(415) 642-5609

Dr. Ghislain de Marsily
Ecole des Mines
Centre d'Informatique Geologique
35 Rue Saint-Honore
77305 Fontainebleau, France
33-6-422-48-21

Dr. Robert O. Fournier
Mail Stop 910
U. S. Geological Survey
345 Middlefield Road
Menlo Park, California 94025
(415) 323-8111, ext. 4158

Dr. Kalman Kovari
Rock Engineering Department
Federal Institute of Technology
ETH-Hoenggerberg, CH-8093
Zurich, Switzerland
01-377-31-82 or 01-377-44-11

Dr. Donald Langmuir
Department of Chemistry
and Geochemistry
Colorado School of Mines
Golden, Colorado 80401
(303) 273-3631

Dr. Chin-Fu Tsang
Earth Sciences Division
Lawrence Berkeley Laboratory
Berkeley, California 94720
(415) 486-5782

Dr. Paul A. Witherspoon
Department of Materials Science
and Mineral Engineering, U. C. B.
and Earth Sciences Division, L. B. L.
Berkeley, California 94720
(415) 486-5082

ACKNOWLEDGEMENTS

The rapporteurs gratefully acknowledge the helpful suggestions and comments of their colleague Chalon Carnahan and the Consultants concerning many aspects of this report.

This report was prepared under the auspices of the Waste Management Branch, Division of Radiation Programs and Earth Sciences, Office of Nuclear Regulatory Research, U.S. Nuclear Regulatory Commission, Washington D.C., 20555 through NRC Fin. No. B3046-3 under NRC Agreement, DOE-60-83-388 and 60-83-367, and the U.S. Department of Energy under Contract No. DE-AC03-76SF00098.

Table of Contents

Table I
Discussion Topics at the Second Consultants' Meeting

Topic	Presented By
Thermohydrological Processes in Unsaturated Porous-Fractured Media	Staff Scientists (Lawrence Berkeley Laboratory)
Sealing of Fractures by Silica Redistribution	G. de Marsily (Ecole des Mines, Paris)
Transport Processes in a Saturated Clay	C. L. Carnahan (Lawrence Berkeley Laboratory)
Thermomechanical and Hydromechanical Processes	J. Noorishad (Lawrence Berkeley Laboratory)
Macrofracture Permeability	B. R. Clark (Leighton and Associates)
Geochemical Processes in Two-Phase Systems	R. O. Fournier (U. S. Geological Survey)
Colloid Transport in Fractured Media & Plans for In Situ Tests of Coupled Processes at Fanay-Augeres	G. de Marsily (Ecole des Mines, Paris)
Review of Coupled Processes Modeling & Geochemical Approaches to Site Characterization and Packing Design	D. Langmuir (Colorado School of Mines)
Field Test Investigations of Coupled Processes in Basalt, Tuff and Salt	LBL Consultants

1. Introduction

The second meeting of the Consultants on Coupled Processes Associated with Geological Disposal of Nuclear Waste occurred on January 15-16, 1985 at Lawrence Berkeley Laboratory (LBL). All the consultants were present except Dr. K. Kovari, who presented comments in writing afterward. This report contains a brief summary of the presentations and discussions from the meeting. The first meeting of the consultants is summarized in an earlier report (LBL Panel, 1984).

The discussion covered the topics listed in Table I. The main points of these topics are briefly summarized below under four major subject areas in sections 2-5. Some points that emerged during the discussions of the presentations are included in the text related to the respective talks.

2. Comments on Coupled Processes in Unsaturated Fractured Porous Media

In the year since the previous meeting of the consultants, scientists at LBL have examined possible significant uncertainties in coupled processes that may affect the performance of a nuclear waste repository. They have found a number of unresolved questions associated with coupled processes occurring in unsaturated fractured porous media. Therefore, LBL staff scientists made presentations to the consultants of their modeling studies of strongly heat-driven flow in partially saturated fractured porous media. They modeled the simultaneous transport of heat, liquid water, vapor, and air. The presence of fractures makes the transport problem very complex, both in terms of flow geometry and physics. There is a need to handle the extreme non-linearities which arise in phase transitions, component disappearances, and capillary discontinuities at fracture faces. From an analysis of the results obtained with explicit fractures, they developed an "equivalent" continuum model that can reproduce the temperature, saturation, and pressure variation, and gas and liquid flow rates of the discrete fractured-porous matrix calculations. This results in a substantial simplification of the flow problem, which makes larger scale modeling of complicated unsaturated fractured porous systems feasible.

The consultants commented that there is a need for such investigations to take into account chemical reactions which may occur. These might have an effect on the permeability, which would affect the flow.

LBL scientists also presented a summary of an analysis of data and a preliminary modeling study of a hydrogeological system in unsaturated media. Flow through the system is controlled by complex interactions between porous media and fractures because some units are highly fractured and others are not. Some field data are available to give profiles of the moisture tension, total potential, degree of saturation and temperature with depth. Although the data are not conclusive, it appears that the average heat flux may be transported by upward flow of water vapor, rather than by conduction alone. Both one- and two-dimensional modeling studies were done to predict the long-term behavior of the system and to investigate the general flow patterns predicted on the basis of available data. Conclusion from the work so far is that lateral flow is likely to be significant in the mainly porous units above the major fractured unit if flow through the fractured unit is controlled by its very low matrix permeability. However, if fracture flow of liquid water is significant, then lateral flow will occur primarily in the mainly porous units below it. The results of these simulations depend greatly on rather poorly known parameters such as the relative permeability curves for unsaturated media and the poorly understood physics of flow through fractured unsaturated media, so the findings must be considered with caution.

3. Comments on Overview of Coupled Processes

Four presentations were made to provide the consultants with an overview of current research in coupled processes. First was a review of work in fully coupled processes by K. Pruess (LBL) concerning redistribution of silica and other minerals, followed by an invited talk by consultant G. de Marsily (Ecole des Mines) on related research in sealing of fractures by silica redistribution. Then a report was given of an investigation of physical and chemical transport processes in a saturated clay by C. L. Carnahan (LBL), and a summary of a survey on thermomechanical and hydromechanical processes by J. Noorishad (LBL).

K. Pruess reviewed processes identified by the consultants and a by literature search which may give rise to significant four-way (T-H-M-C) coupling under conditions expected near nuclear waste repositories. In particular, the redistribution of rock minerals with associated porosity and permeability changes has been investigated. Silica is the best known and probably most important mineral to be redistributed in the context of nuclear waste isolation. Redistribution of silica has been extensively documented and studied by many researchers as an important effect in natural hydrothermal convection systems. Much basic information about the chemistry involved in silica dissolution and precipitation is presently available. Quantitative methods for predicting the impact of these effects on hydrothermal systems are as yet rather undeveloped. However, it appears that a meaningful evaluation of potential effects from silica redistribution on the performance of a nuclear waste repository is possible with existing methods, at least in a generic sense. A specific issue which should be addressed in future research is whether silica redistribution might have the potential of altering the evolution of a repository system to such an extent that large pore pressure buildup with subsequent hydrofracturing might occur. This possibility would be troublesome, as it may lead to a serious breach of containment.

Until such studies have been completed it is not possible to anticipate whether silica redistribution will have a significant or a negligible impact on repository performance. The answers may well depend upon site-specific conditions, and may change when different temporal and spatial scales are considered. Pruess suggested that consideration of possible redistribution effects from the minerals be postponed until the silica problem has been analyzed in reasonable detail, and the likelihood for significant effects has been established.

At the invitation of the consultants, G. de Marsily presented a summary of a study of the sealing of fractures by silica by A. Ribstein of the Ecole des Mines, Paris. She employed a single-fracture model with time-varying thermal and velocity gradients. The solubility of amorphous silica was used with first order reaction kinetics. An assumption was made that the aperture of the fracture remained constant as silica precipitated, in order to preserve a constant flow. She found that concentration of silica in solution depended only on temperature, not on the velocity or rate constant. Silica deposition amounted to approximately 23% of the fracture aperture in 10,000 years, which indicates that sealing of fractures by amorphous silica may not be significant. The highest temperature considered was 70 °C.

A review of coupled physical and chemical effects in the packing surrounding a waste canister was presented by C. L. Carnahan of LBL. In near-field regions of nuclear waste repositories, thermodynamically coupled flows of heat and matter can occur in addition to the independent flows in the presence of gradients of temperature, hydraulic potential, and composition. The following coupled effects can occur: thermal osmosis, thermal diffusion, chemical osmosis, thermal filtration, diffusion thermal effect, ultrafiltration, and coupled diffusion. Flows of heat and matter associated with these effects can modify the flows predictable from the direct effects, which are expressed by Fourier's law, Darcy's law, and Fick's law. The coupled effects can be treated quantitatively together with the direct effects by the method of the thermodynamics of irreversible processes. The extent of departure of fully coupled flows from predictions based only on consideration of direct effects depends on the strengths of the gradients driving flows, and may be significant at early times in clay packing materials and in near-field geologic environments of repositories. Approximate calculations using data from the literature and reasonable assumptions of repository conditions indicate the thermal-osmotic and chemical-osmotic flows of water and solutes in semipermeable packing may exceed flows predicted by advection and diffusion alone by two to three orders of magnitude. In permeable materials, thermal diffusion may contribute to solute flows to a smaller, but still significant, extent.

These considerations imply needs for further experimental and computational research. Characterization of materials proposed for use as packing should include measurement of phenomenological coefficients associated with thermodynamically coupled processes; this is especially important if the saturated materials can be expected to behave as semipermeable membranes. Also, numerical simulators of transport processes which are to be used to evaluate the performance of proposed nuclear waste repositories should be expanded in scope to include possible thermodynamically coupled processes in the near-field region. Sensitivity calculations should be done to evaluate the significance of thermodynamically coupled effects in relation to the direct effects.

J. Noorishad of LBL reviewed research in the thermomechanical and hydromechanical processes. The research is concerned with three key issues. The first issue is to find the research works aimed at understanding the phenomena occurring at both the microscopic and macroscopic scales in the host rock mass. The second is to survey the modeling techniques and efforts that can realistically simulate the coupled processes at various levels of the repository environment over time. The third area is to search for data characteristic of the full range of variation of conditions in the life of the repository, and to survey the state-of-the-art methods and techniques of laboratory and field measurements of rock and rock mass properties. To understand the thermomechanical and thermohydromechanical processes in a rock mass one needs an adequate description of the effects at microscopic or crystalline levels. The events which occur and are observable in the larger scale in the repository include fracture permeability development, spalling and cave-ins. Contributing factors to these effects include microcracking, macrocracking, recrystallization and thermodynamic instability.

The numerical modeling of these coupled processes needs the input of geometry, kinematics (strain/displacement) and, most important, the constitutive model. A list of thirteen important constitutive models was given from which an adequate numerical model must draw. The computational needs of such a model may be met by a promising scheme that is a hybrid of the finite element method and the boundary element method.

For predictive modeling studies of a geotechnical structure a nine-step methodology was given based on two basic elements, site characterization (including laboratory and field measurements) and advanced computational methods. The state of the art for laboratory and in situ field testing and measurements was highlighted in a table, "Field Measurement Data", which listed details of the needed rock mass properties and the instruments currently available to measure them. Rock properties were listed in a table that covered the lithologic, physical, and geophysical properties in addition to chemical properties that need adequate measurement. This research showed that the current understanding of coupled thermomechanical and thermohydromechanical processes needs further development and additional data from measurements. As these increase, site characterization and predictive modeling to establish criteria for assessing potential repository performance should be feasible.

4. Presentations by Consultants on Selected Topics of Current Interest in Coupled Processes

Four consultants gave talks on topics in their special expertise related to coupled processes associated with nuclear waste repositories: B. Clark, R. O. Fournier, G. de Marsily and D. Langmuir. Their presentations are summarized in a brief form here.

B. Clark reviewed the possibility of macrofractures acting as large permeability channels at a site. When large fractures or groups of fractures develop, matrix permeability will be negligible in comparison, and this could happen at any site. There is evidence in various areas of engineering literature for large fractures remaining open for long periods of time, and also for their healing. Fractures remain open as a function of fluid and rock properties, the stress field, and the fracture geometry (especially fracture lengths). There is available a number of investigations in petroleum engineering and geothermal research on these topics, e.g., the hot dry rock experiment. Also, the research on ore deposits may provide valuable information. The case study of hydrothermal tin deposits at Panasqueira, Portugal (Kelley and Rye, 1979) was given as an example where wide horizontal joints were held open for a long time simultaneously. This indicates that we need to analyze worst-case models and do sensitivity studies in order to set limits on conditions at a site.

A report of investigations of boiling phenomena in the hydrothermal system at Yellowstone National Park was given by R. O. Fournier. There have been detailed records of chloride and other constituents in the springs and geysers at Norris in Yellowstone for 25 years. Over this time an annual variation was noted for chloride and sulfate ions. Investigations by Fournier and others of the mechanism for these variations developed the following possible explanation: the drop of the water table in the fall produces boiling in the hydrothermal reservoirs, and a simultaneous drop in fluid pressure allows mixing of three types of waters. When the vapor condenses, there is an acid attack on the substances where it condenses. HCl and other species could be transported through the packing if there were no condensation in the packing.

In experimental work at the vapor pressure of the solution, Fournier has shown that feldspar is dissolved in the liquid and kaolinite deposited on quartz suspended in the vapor phase, implying the transport of aluminum in the vapor phase. The total mass flux in vapor transport may be negligibly small.

G. de Marsily made a presentation of two topics of current research in France: colloid transport and plans for in situ experiments for coupled processes. Colloid transport is not important in porous media but it is in fractured systems. A colloid with the same charge as the surface of the fracture will be repelled by the surface, and the velocity of the particle may be from 1.1 to 1.4 times the velocity of the water, producing hydrodynamic chromatography. G. de Marsily did an investigation to review a KBS (Swedish) study which said that uranium colloids would be formed near the canister, and as they migrate along a fracture with uranium in solution, the solution will diffuse into the porous matrix causing the colloids to redissolve. De Marsily formulated a model which also included the sorption of colloids on the fracture walls and sorption of uranium by the porous matrix. If colloid sorption were negligible, the colloids would reach the outlet in only 100 years. If there were no colloids, the uranium in solution would take 1 million years to reach the outlet. This demonstrates that in fractured systems, colloids may be important, and their properties should be measured experimentally in the laboratory.

A report was made also of plans for in situ experiments on coupled processes in a uranium mine in granite at Fanay-Augeres in France. Preparations are being made for a series of experiments similar to the ventilation experiment at Stripa, Sweden, with a number of packed-off boreholes radiating from the end of a drift. There are plans for a series of hydraulic tests to ascertain local permeabilities in the fractured rock system, and tracer tests to discover the mass transport properties. Also coupled thermohydrological experiments have been designed with heat power and geometry scaled to give a time scale reduction of 100,000 years to 1 year for a scale model of the repository. The fracture system is being characterized with the aid of geostatistical methods. They have found that to effectively apply geostatistical techniques to fracture parameters such as length and density, they must apply them to particular families of fractures. Through the experiments and the geostatistical characterization of the rock mass, they hope to estimate the global permeability.

D. Langmuir presented three main topics concerning the application of geochemical concepts to coupled processes associated with a nuclear waste repository. The first was a summary of a paper by F. J. Pearson, Jr. (1984) concerning the linking of flow and geochemical models. The second and third related new ideas for geochemical approaches to site characterization and packing design respectively. Langmuir also briefly related the current status of the NEA-CODATA project to provide an internally consistent thermodynamic data base for the actinides, including data for complexes and minerals.

The current status of coupling geochemical and solute transport models has been comprehensively reviewed by Pearson (1984). Among strictly geochemical models, most assume chemical equilibrium exists. All such models can calculate the distribution of species in the aqueous phase (homogeneous reactions). Many assess reactions involving also associated mineral species and a gas phase (heterogeneous reactions). The heterogeneous reactions involve "mass transfer" (cf. Nordstrom and Ball, 1984).

The geochemical models (whether or not coupled with mass transport) require as input information: (1) an accurate chemical analysis of the water expressed as total species concentrations, or as activities or activity ratios when electrodes are used for measurement; (2) the chemical and mineralogical analyses of the rock, and/or gas phase when heterogeneous reactions are considered; and in every case, (3) a reliable thermodynamic data base. Such data bases are generally fair to good for major minerals and dissolved species as a function of temperature, pressure and ionic strength, but fair to poor for most radionuclides. The output from such codes is: (1) the activities of the ions and complexes; and (2) the saturation state of the waters with respect to the minerals (data on solid solutions is lacking).

The geochemical models can also be described as considering conditions in a static water or water/rock system at equilibrium, or considering reaction progress, that is, states of partial equilibrium in the path towards complete equilibrium. Well-known examples of static models are WATEQF (Plummer and others, 1976) and SOLMNEQ (Kharaka and Barnes, 1973). Reaction progress codes include PHREEQE (Parkhurst and others, 1980) and EQ3/EQ6 (Wolery, 1983).

Most approaches to the problem of describing migration of chemicals in flowing ground water can be called "solute transport models". These do not represent a true coupling of chemical reactions with flow. Thus, certain models of this type exclude all chemical reactions and simulate only the effects of advection and hydrodynamic dispersion on solute concentrations (Konikow and Bredehoeft, 1978). In other models, concentrations in the fluid phase are related to composition of sorbed phases by simple empirical expressions called isotherms (Van Genuchten and Cleary, 1979). In general, the only chemical interactions included in solute transport models are first-order production and decay of radionuclides and equilibrium-controlled sorption or idealized ion-exchange reactions. These models do not account for effects of chemical changes on fluid flow. The simplest coupled solute transport-geochemical codes consider only a few of the possible geochemical reactions expected in the water/rock system. Such codes may incorporate: (1) radioactive decay; (2) sorption, as described by ion exchange or with linear isotherms such as K_d; (3) equilibrium controlled mineral precipitation/dissolution (no kinetics). However, they often use aqueous concentrations rather than activities, ignore complexes, and in general assume fluid flow is independent of chemistry. A sophisticated coupled code which answers most of these criticisms, CHEMTRN (Miller and Benson, 1983), is discussed below.

There are two main approaches to coupling the geochemistry and solute transport models: direct and iterative coupling. In the direct coupling approach, transport equations are written for each basis species, and other species are related to the basis species by mass action relations. Examples of this approach are the models of Rubin and James (1973) and Valocchi and others (1981). However, both models employ concentrations rather than thermodynamic activities in their mass action relations, and neither model includes complex ion formation or precipitation/dissolution of solid phases. A more rigorous approach is taken by the CHEMTRN code (Miller and Benson, 1983), which simulates ion exchange, includes the surface complexation ionization model--the best available model for calculating adsorption of trace elements--and precipitation/dissolution. The CHEMTRN code employs thermodynamic activities rather than concentrations. (See also Jennings and others (1982).) In the iterative approach the flow model moves the species to a point in a volume where it is reacted with other species by the geochemical model. Examples of this kind of coupling are found in Grove and Wood (1979) and Walsh and others (1982). The latter authors also employ the surface ionization and complexation model. They investigate the dissolution of rock by acid, calculate a porosity change, and from this compute a permeability change. None of these models include kinetics of chemical reactions in their calculations.

Pearson (1984) summarizes his review with comments on the calculation of the effects of chemistry on flow and matrix properties. The chemistry affects these properties in three ways: (1) permeability and porosity of the rock matrix, (2) viscosity and density of the fluid, (3) fluid pressure (osmotic). No models consider porosity and permeability effects directly, instead using empirical or theoretical relations between porosity and permeability. Pearson is wrong in saying there is a general relation between porosity and permeability. The density may be a function of salinity, pressure and temperature, and the viscosity a function of salinity and temperature. The osmotic fluid pressure may be of interest only in the near field. It was pointed out in discussion that these models do not consider boiling, but it is important, changing salinity and affecting the flow model.

In site characterization, geochemical data available from proposed sites is being inadequately utilized. At least five types of such data provide invaluable and unique information as to site suitability. First, we can use water/rock reaction models such as WATEQF (Plummer and others, 1976) or PHREEQE (Parkhurst and others, 1980) on chemical analyses of water samples from exploratory boreholes. Such information from some of the Swedish granite ground waters and Texas Palo Duro Basin brines suggests that some of the waters were saturated with respect to forms of UO_2 and ThO_2. This indicates that if uranium (or thorium) were released to such ground waters from a repository, it would tend to precipitate rather than be adsorbed, and that maximum uranium concentrations in case of a breach would probably be controlled by UO_2 precipitation, and so can be predicted. In this case, sorption K_d's would be meaningless.

Second, we can study the radioactivity of daughter products of natural ^{238}U and ^{232}Th in the ground water. For example, equal radioactivities of ^{226}Ra and daughter ^{222}Rn, an inert gas which is not adsorbed, indicates that the ^{226}Ra is also not adsorbed (Laul and others, 1985). Equal radioactivities of ^{234}U and daughter ^{230}Th, suggest that uranium in the ground water is in the reduced 4+ valence state. Equal activities of ^{228}Ra and ^{224}Ra indicate that intermediate ^{228}Th is adsorbed in the present water/rock system (cf. Laul and others, 1985).

A third under-utilized geochemical approach to site characterization is to age date the ground water. If one finds ^{14}C (half-life 5570 years) or ^{3}H (tritium; half-life 12.26 years), the site is not adequately isolated. Other isotopes, such as ^{36}Cl, and the isotopes of He, Ar, Kr, and U can be used for dating, although these require data and assumptions regarding water/rock ratios.

Fourth, stable isotope ratios such as those involving hydrogen and deuterium, ^{18}O and ^{16}O, ^{13}C and ^{12}C, and ^{34}S and ^{32}S can be used to identify the source or sources of the ground water, to reconstruct its evolution, and even to reconstruct the temperature of the water as initial rainfall.

Fifth, the inert gases He, Ar, Xe, Kr have solubilities which are functions of temperature and salinity. Among these gases, the concentrations and ratios of those not also derived from radioactive decay (He and Ar) provide information as to the original temperature and salinity of the water in contact with the atmosphere.

A final matter to consider in the application of geochemistry to coupled processes in nuclear waste isolation is the chemical design of the packing surrounding the waste canister. This design has three objectives: (1) to minimize the flow of groundwater, (2) to maximize the attenuation of radionuclides, and (3) to ensure structural stability over the long term. From this point of view we can evaluate proposed packing substances, and consider mixtures of compounds or additives to achieve the desirable properties. For example, bentonite has positive characteristics of good swelling properties, low permeability and high cation exchange capacity, but has negative characteristics of instability at higher temperatures and no redox control. (It was remarked in the discussion that erosion of bentonite is a source of colloids.) For another example, zeolites have high cation exchange capacity, particle sizes larger than clay size, and good thermal stability, but still do not exert redox control.

Controlling the oxidation state of the repository is important in that the chief element in spent fuel is uranium as UO_2, which is highly insoluble under reducing conditions. To achieve redox control, we can introduce an additive to the packing such as pyrite (FeS_2), or ilmenite ($FeTiO_3$) which has been sulfidized. Thus treating ilmenite with

H_2S yields FeS_2 to produce reducing conditions, and relatively amorphous TiO_2 which is perhaps the strongest inorganic sorbent known (cf. Langmuir, 1978). From a cost standpoint, all the minerals just described are available in large quantities at reasonable prices.

Another approach to packing design is to use a packing that will react at repository temperatures to form a single crystal or at least non-porous crystalline phase. For example, a stoichiometric mixture of crushed iron-rich olivine and diatomaceous earth in a water slurry (the Fe^{2+} exerts redox control) will form serpentine and seal the canister. The serpentine, which would be stable under repository conditions, would create a low permeability environment.

Gypsum or anhydrite and calcite have retrograde solubilities with temperature, and can be expected to precipitate near or at the hot canister. On the other hand, the solubility of silica decreases with decreasing temperature, so that silica will dissolve near the waste and precipitate further away from it. These minerals can easily be added to the packing if not already present in the rocks. The result would be a large reduction in porosity and permeability of the packing and adjacent country rock caused by clogging with mineral precipitates.

Concern has been expressed regarding the production of mobile uranyl (UO_2^{2+}) species by radiolysis. To counter this possibility, we can add phosphate or vanadate species (or P or V oxides) to the packing. In the presence of natural levels of K^+ and Ca^{2+} very insoluble uranyl phosphate and vanadate minerals are precipitated. In this context, J. Cramer (1985, AECL, Pinawa, Manitoba, personal communication) reports that is is very easy to precipitate the calcium uranyl vanadate, tyuyamunite, in the laboratory at 150 °C using the calcium-chloride brines of the Canadian Shield.

If migration of uranium from the waste as U^{4+} is possible, we can add ore-grade UO_2 to the packing. By establishing a chemical potential gradient for uranium towards the canister, we can minimize or reverse migration of uranium from the waste package.

Finally, if a high salt content is added to the packing to produce more saline waters near the waste than present in the country rock, one can create an osmotic flow back towards the canister.

5. Recommendations for Underground Field Tests with Applications to Three Geologic Environments

The consultants closed their deliberations with a consideration of what tests of coupled processes they would recommend to be performed in an underground field test facility. After some discussion, it was decided that certain basic tests should be performed, with variations to suit the particular geologic environment and host rock type as appropriate. The recommended tests are listed below with general comments for each one. There follow specific comments concerning the performing of such tests in three rock types widely considered as potential hosts for a nuclear waste repository: basalt, tuff and salt.

There were seven basic tests recommended:

(1) Site Background Characterization

This includes measurements of the thermal, hydrologic, mechanical and chemical properties and conditions at the site as background data before any tests are conducted. It would incorporate the geologic description of the host rock, including (where relevant) the fracture system.

(2) Monitoring of Fractures During Excavation

The excavation of shafts, tunnels and chambers of a repository will likely induce new cracks or cause existing joints to be opened in the vicinity of these constructions. Large scale continuous strain measurements during the advance of the excavation of these structures will provide information along the underground system. High precision strain distribution monitoring using the borehole sliding micrometer could answer the question whether nuclides can reach the accessible environment faster along the man-made structures than through the undistrubed rock formations, and contribute to studies of hydromechanical (H-M) couplings (NAGRA, 1985).

(3) Permeability Megatest

This test would be on the scale of hundreds of meters, such as the ventilation experiment at Stripa Mine, Sweden. It might include many instrumented boreholes which could also be utilized for tracer tests.

(4) Thermohydromechanical (T-H-M) Perturbation Test

This test would also be on the scale of hundreds of meters. It would mean monitoring the response of the rock to the creation of a hydraulic sink (M-H), and to the effect of a heat source (T-M), and then to a pressurized sealed heat source (T-H-M) which could be allowed to cool down while saturated to see the coupled effects.

(5) Thermohydrochemical (T-H-C) Migration Studies in the Rock Matrix

This test would be on the scale of a few meters to investigate the transport properties of the host medium to both tracers and colloids.

(6) Thermohydrochemical (T-H-C) Migration Studies in Proposed Packing

These experiments should also be on the near canister scale to examine the behavior in situ of the proposed packing to various conditions. Certain parameters such as Eh (redox potential) may be tested for the sensitivity of the packing to changing conditions over time.

(7) Site-Specific Tests

These would include investigations of characteristics which are unique to a particular rock type or host environment. They would include such tests as examining creep behavior and fracture healing in salt, and water infiltration in unsaturated tuff.

(8) Mine Back

This is excavation of a tested portion of the host rock to examine in detail the effects of tracer migration, hydrothermal changes, proportions of tracer in pores and on fracture surfaces, etc.

In applying these tests to basalt, the consultants discussed in particular examining the mechanical response to the drained condition and then to a pressurized condition. Vertical permeability should also be examined.

In unsaturated tuff the permeability megatests may need to be done as air pressurized tests, and heated-air pressurized tests. Also, experiments can be done with saturated air to investigate vaporization and condensation, with sampling and analysis of the condensate afterwards. Water infiltration experiments and an investigation of thermal effects on moisture content should also be done. For tracer studies, gas phase tracers may need to be employed.

For salt, a megascopic test in low permeability environments may need to be designed. Permeability to brine flow should be examined. Tests need to be made to investigate whether there is flow in fractures that do not heal. Fracture healing and salt creep rates should be studied.

References

Grove, D. B., and Wood, W. W., 1979, Prediction and Field Verification of Subsurface-Water Quality Changes during Artificial Recharge, Lubbock, Texas, Ground Water, 17(3), 250-257.

Jennings, A. A., Kirkner, D. J., and Theis, T. L., 1982, Multicomponent Equilibrium Chemistry in Groundwater Quality Models, Water Resources Research, 18(4), 1089-1096.

Kelley, W. C., and Rye, R. O., 1979, Geologic, Fluid Inclusion, and Stable Isotope Studies of the Tin-Tungsten Deposits of Panasqueira, Portugal, Economic Geology, 74(8), 1721-1822.

Konikow, L. F., and Bredehoeft, J. D., 1978, Computer Model of Two-Dimensional Solute Transport and Dispersion in Groundwater, Techniques of Water-Resources Investigations of the U. S. Geological Survey, Book 7, Ch. C2, 90 pp.

Langmuir, D., 1978, Uranium Solution-Mineral Equilibria at Low Temperatures with Applications to Sedimentary Ore Deposits, Geochimica and Cosmochimica Acta, 42, 547-579.

LBL Panel, 1984, Panel Report on Coupled Thermo-Mechanical-Hydro-Chemical Processes Associated with a Nuclear Waste Repository, C. F. Tsang and D. C. Mangold, eds., Lawrence Berkeley Laboratory Report LBL-18250.

Miller, C. W., and Benson, L. V., 1983, Simulation of Solute Transport in a Chemically Reactive Heterogeneous System: Model Development and Application, Water Resources Research, 19(2), 381-391.

NAGRA Underground Rock Laboratory Project, 1985, "Loosening Zone" Report on Monitoring and Interpretation of Results.

Nordstrom, D. K., and Ball, 1984, chapter *in* C. J. M. Kramer and J. C. Duinker, eds., Complexation of Trace Metals in Natural Waters, M. Nijhoff and W. J. Junk Publishers, 149-164.

Parkhurst, D. L., Thorstenson, D. C., and Plummer, L. N., 1980, PHREEQE: A Computer Program for Geochemical Calculations, U. S. Geological Survey, Water Resources Investigations 80-96.

Pearson, F. J., Jr., 1984, Linking Flow and Geochemical Models: Status and Problems of Developing a Fully Coupled Model, in The Geochemistry of High-Level Waste Disposal in Granitic Rocks, AECL/CEC Workshop, Sept. 12-16, 1983, Minster Louel, UK, N. A. Chapman and F. P. Sargent, eds., publ. by AECL/WNRE and CEC.

Plummer, L. N., and others, 1976, WATEQF - A Fortran IV Version of WATEQ, A Computer Program for Calculating Chemical Equilibrium of Natural Waters, U. S. Geological Survey Water Resources Investigations 76-13.

Rubin, J., and James, R. V., 1973, Dispersion-Affected Transport of Reacting Solutes in Saturated Porous Media: Galerkin Method Applied to Equilibrium-Controlled Exchange in Unidirectional Steady Water Flow, Water Resources Research, 9(5), 1332-1356.

Valocchi, A. J., Street, R. L., and Roberts, P. V., 1981, Transport of Ion-Exchanging Solutes in Groundwater: Chromatographic Theory and Field Simulation, Water Resources Research, 17(5), 1517-1527.

Van Genuchten, M. Th., and Cleary, R. W., 1979, Movements of Solutes in Soil: Computer-Simulated and Laboratory Results, *in* G. H. Bolt, ed., Soil Chemistry B: Physico-Chemical Models, Elsevier Scientific Publishing Co., Amsterdam, 349-386.

Walsh, M. P., Lake, L. W., and Schecter, R. S., 1982, A Description of Chemical Precipitation Mechanisms and Their Role in Formation Damage during Stimulation by Hydrofluoric Acid, paper presented at 1982 International Symposium on Oilfield and Geothermal Chemistry, Society of Petroleum Engineers, Dallas, Texas, Jan. 25-27, 1982.

Wolery, T. J., 1983, EQ3NR: A Computer Program for Geochemical Aqueous Speciation-Solubility Calculations: A User's Guide and Documentation, University of California Report UCRL-53414.