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WASTE-ISOLATION PROJECTS FY 1978

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WASTE-ISOLATION PROJECTS: FY 1978

ABSTRACT

This report describes Lawrence Livermore Laboratory (LLL) activities during FY 1978 in support of the National Waste Terminal Storage Program. Current projects at LLL fall into three categories: (1) field testing, (2) laboratory rock mechanics measurements, and (3) laboratory studies of sorption and leaching.

Field test activities conducted in the Climax granite at the Nevada Test Site included electrical heater tests, preparation for a spent-fuel-storage test, and planning for a series of rock mechanics tests. The heater tests determined the *in situ* thermal properties of Climax granite and its *in situ* permeability as a function of rock temperature.

The two main laboratory rock mechanics projects involved (1) measurement of the permeability, electrical conductivity, and acoustic velocity of 15-cm-diam cores of granitic rocks over a range of confining pressure, pore (water) pressure, and deviatoric stress, and (2) measurement of rock thermal properties as a function of temperature and confining pressure in the presence of pore fluids to 770 K and 200 Mpa.

The leaching studies made use of an LLL-designed, single-pass leaching apparatus with three solutions, two leach temperatures, and three flow rates. The material evaluated was Np-Pu-doped simulated waste glass from Battelle Pacific Northwest Laboratories. The sorption studies involved standard static measurements of the equilibrium distribution coefficient (K_d) for various radionuclides on a variety of rocks, and flow-through-core studies of dynamic sorption.

1. INTRODUCTION

1.1 LLL Program Goals

During FY 1978, the primary goal of the LLL waste-isolation program was to develop the technology and data base necessary to license a nuclear waste repository in a crystalline rock medium, with emphasis on a location at or near the Nevada Test Site (NTS). Our secondary goal was to assist DOE (within areas of technical strength at LLL) to develop repositories in other rock types and locations.

1.2 Technical Approach

The primary emphasis of the LLL program is on developing a nuclear waste repository in granitic rock. There are distinct advantages to having such a program. Of the states that lack salt or flood basalt formations, 20 have granitic rocks. In fact, only 5 of the 50 states are entirely without granitic rocks between the ground surface and 900 m below sea level.

Thus, the validation of granite as a repository medium will greatly expand location options for nuclear waste repositories.

Another advantage of crystalline igneous rocks in general, and of granite in particular, is the fact that they offer a superior potential for retrievability. Even under thermal loads from waste emplacement, openings in granitic rock can be designed for 50- to 100-year stability. Retrievability is important for added public assurance and because it allows the option of planned intermediate storage.

The low ionic strength of the ground water associated with granitic rocks makes the use of engineered migration barriers such as specially formulated waste forms, special canister materials, and backfill more feasible than in salt. There is a general consensus that fission products decay to trivial levels in the 600- to 1000-year time-frame, and it is quite conceivable that we can contain the waste within engineered barriers for such times.

Another advantage of pursuing a granitic waste repository option in the United States is the opportunity to utilize information from three technically active foreign programs in granitic rock: a vigorous and advanced program in Sweden and research programs in Canada and Great Britain. The Swedish Program¹ has produced an entire waste management system based on a granitic rock disposal medium. Field rock mechanics tests are going on in Sweden² and in England,³ and they are planned in Canada.⁴ In Sweden, field tests of the migration of radionuclides have been carried out,⁵ and more are planned. By building on results from these foreign programs, a U.S. program can advance more rapidly and at a lower cost than would otherwise be possible.

At a secondary level of emphasis, we have carried out laboratory measurements in geochemistry and rock mechanics on rock media other than granitic rocks. These efforts are compatible with our overall goals.

1.3 Description of the Project

Current projects at ILL fall into three categories: (1) field testing (including mathematical modeling), (2) laboratory rock mechanics measurements, and (3) laboratory studies of sorption and leaching. Work in all three of these categories was ongoing in FY 1977 and was summarized in last year's annual report.⁶

1.3.1 Field Testing

During FY 1978, all field test activities involved the Climax granitic stock (Climax granite) at NTS, an excellent location for generic tests. There are existing surface facilities, utilities, a vertical shaft (2×3 m) for access to excavations 420 m below ground surface, and a large amount of background technical information obtained during previous tests. The facility is government-owned, and radioactive material is already in "storage" from previous nuclear test activities.

Field activities during FY 1978 included heater tests, preparation for a spent-fuel-storage test, and planning for a series of rock mechanics tests.

The heater test was initially carried out to measure the thermal properties of Climax granite *in situ* and thus consisted of thermal measurements only (see Sec. 2.1.1). Following completion of the thermal measurements, one heater and four thermocouple assemblies were removed and reconfigured to make repeated remotely controlled transient permeability measurements as a function of rock temperatures (Sec. 2.1.2).

As early as FY 1976, there was interest in the test storage of simulated or actual reactor waste in the Climax granite. In June 1978 we received funding for a spent-fuel-storage test in the Climax granite (Sec. 2.2). The test layout will be constructed in FY 1979, and operation will start in FY 1980.

Throughout FY 1978, planning continued for a series of rock mechanics tests in the Climax granite (Sec. 2.3). These tests will follow the heater tests and complement the spent-fuel test.

1.3.2 Laboratory Rock Mechanics Measurements

The basic goal of our laboratory rock mechanics program has been to extend measurement capabilities so as to obtain currently unavailable data necessary for analysis of the effects of waste emplacement in rock.

There are two main projects. The first (Sec. 3.1) involves measuring permeability and other parameters on large (15-cm-diam) specimens over a range of confining pressures, pore (water) pressures, and deviatoric stresses. These systematic data can be used to analyze the effect of depth on rock permeability. During the same test loading, permeability, stress, strain, electrical conductivity, and acoustic velocity are simultaneously measured. If electrical conductivity or acoustic velocity can be shown to correlate with permeability, then a remote diagnostic tool will be indicated.

The second main project (Sec. 3.2) involves measuring rock thermal properties as a function of varying temperature and confining pressure in the presence of pore fluid to 770 K and 200 MPa. Such data are needed for predictive calculations of the effects of waste emplacement. The thermal parameters measured are conductivity, diffusivity, heat capacity, and the linear expansion coefficient.

There have been several other projects involving the measurement of permeability and thermal properties. They are summarized in Secs. 3.3 and 5.1.

1.3.3 Laboratory Studies of Leaching and Sorption

The laboratory studies of leaching and sorption are part of a Waste Isolation Safety Assessment Program (WISAP) at the Battelle Pacific Northwest Laboratories (PNL). The leaching activities, which provide data on the availability of radionuclides in waste to flowing ground water, are part of WISAP Task 2, Waste Form and Characterization. The

sorption studies, which provide data on the removal or retardation of radionuclides from flowing ground water, are part of WISAP Task 4, Nuclide Transport Data.

During FY 1978, the leaching studies (Sec. 4.1) were carried out by means of an LLL-designed single-pass leaching apparatus using Np-Pu-doped simulated waste glass supplied by PNL. We are doing dynamic (one-pass) leaching of doped simulated waste glass to compare with the PNL static (IAEA or Hespel) leaching of similar glass. PNL is also leaching actual waste glass in a hot cell by the

same method for comparison with their doped-glass experiment. Thus, the LLL-PNL pair will compare dynamic versus static leach method, and the PNL-PNL pair will compare static leaching of simulated versus real waste glass.

The sorption studies (Sec. 4.2) consisted of both standard static measurements of the equilibrium distribution coefficient (K_d) for various radionuclides on a variety of rocks, and flow-through-core studies for dynamic sorption.

L. Ramspott

2. FIELD TESTING

2.1 Heater Tests

In the nine-month period from October 1977 to July 1978, three *in situ* tests were conducted in the Climax granite at the Nevada Test Site. The first two tests determined the thermal conductivity and diffusivity of the rock mass in two orthogonal directions. The third test measured the *in situ* permeability of the granite during a thermal cycle. Data analyses and report preparation were in progress at the end of FY 1978.

At the end of FY 1977 the field construction for the heater tests had been completed, and installation of the experimental apparatus was in progress. The experimental array consisted of AX (48-mm-diam) and NX (76-mm-diam) diamond-cored vertical holes arranged in an orthogonal layout (Fig. 1). The holes are 9 to 12 m deep, and the midplane of the instrumentation is located 7.5 m below the drift floor at a depth of 425 m below the ground surface.

2.1.1 Thermal Measurements

Instrumentation Installation. The instrumentation for the heater tests consisted of power controllers and transducers for the heaters and an array of sheathed Type K (Chromel-Alumel) thermocouples. The heaters were powered by phase-fired SCR controllers and monitored by dc-output watt transducers. The thermocouples were installed in groups of four per hole and typically located with two active junctions at the midplane. The thermocouples were interfaced to the recording system at a 60-channel reference junction block located in the experimental alcove. The μ V-level outputs were transmitted to the surface-located recording system on shielded multi-conductor cables.

The installation of the instrumentation array was completed on 13 October 1977, and all heater and instrument holes were backfilled with vermiculite insulation from the top of the instrument array to the floor of the drift.

Heater Test Operations. The first heater test, designated H-1, involved the heater in the alcove and was operated for a total of 68 days from 14 October to 21 December 1977. During this test, power levels and time durations were selected to achieve a constant average power of 3.7 kW. Two related unanticipated events occurred during this test:

- Small quantities of water, probably circulating fluid lost to the fracture system during the drilling operation, migrated into the heater hole as vapor and condensed above the heater.

- The water apparently refluxed in the region near the top of the heater and caused a short in the heater power leads.

The refluxing of water resulted in a distortion of the thermal field, with slightly-higher-than-anticipated temperatures observed above the midplane of the array. The electrical short was readily repaired, and the connection was modified to mechanically isolate it from the wet region.

The second heater test, designated H-2, involved the heater in the drift and was operated for a period of 62 days from 21 December 1977 to 21 February 1978. This test was also operated with power levels and time durations to achieve a constant average power of 3.7 kW to compare with the results of the first test. An excursion of the heater temperature was noted during a high-power (4.8 kW) phase of the test. This excursion was subsequently associated with the accumulation of fine granite dust (probably

FIG. 1. Plan view of heater test No. 1 layout, 420 m level, Climax granite (note that location dimensions for drift holes are the same as those for alcove holes).

drill cuttings) at the bottom of the heater and with a resultant loss of heat dissipation in that region. Similar water refluxing was observed in the region above the heater during this test.

Heater Test Results. The two heater test sequences indicate that the thermal conductivity of the experimental site is essentially isotropic and only slightly temperature-dependent over the range of temperature investigated. The peak rock temperature achieved was approximately 350°C . The thermal conductivity derived from the experimental data

is $3.0 \text{ W/m}\cdot\text{K}$, about 15% higher than the mean value of laboratory measurements without confining pressures. The derived thermal diffusivity is $1.2 \text{ mm}^2/\text{sec}$ and is in good agreement with laboratory measurements on intact core samples. Figure 2 shows a computer plot of the temperature increases as a function of time at four different ranges from the H-2 heater. Note the steeply sloping inflection about 400 h into the test. This corresponds to a brief "power-off" period. Detailed analysis of this inflection permits us to derive the thermal diffusivity.

FIG. 2. Comparison of thermal history at various distances, Test H-2.

2.1.2 Permeability Measurements

Instrumentation Installation. Following the H-2 heater test, the cool-down history was monitored for 10 days. The borehole instrumentation in five holes in the proximity of the H-2 heater was then reconfigured to conduct a set of *in situ* permeability tests at elevated temperatures.

The new instrumentation, shown schematically in Fig. 3, consisted of inflatable rubber borehole packers with pressure-tight feedthrough connections to permit thermocouples and power leads to penetrate the region below the packer. A manifold with precision pressure transducers and solenoid valves enabled us to pressurize the boreholes individually with air and monitor the pressures from the surface and to record all the relevant data, utilizing the same recording system as used for the heater tests. The thermocouple support frames were redesigned to permit their operation in conjunction with the packers.

The revised instrumentation installation, calibration, and checkout procedures were completed on 13 April 1978.

Permeability Test Operations. The permeability test ran for 83 days from 13 April to 7 July 1978. It included a total of seven pressurization test cycles, six of which yielded interpretable results. During the calibration and checkout operation, it was established that, within the resolution of the instrumentation, there was no pressurized air communication between any of the holes in the instrument array. We therefore concluded that the permeability could be obtained by air-pressurizing each hole instantaneously and observing the pressure decay with time. Also, during the checkout operation, it was determined that the time required for significant pressure decay was of the order of several days. Typically, during a 100-h period, the pressure would decline from 750 kPa to 700 to 720 kPa. The pressure-transducer recording resolution was 0.05 kPa.

FIG. 3. Schematic of measurement system for permeability test.

Ideally, there would be no temperature change during a permeability test, so that all pressure change would be due to fluid flow away from the drill hole. Attaining a constant rock temperature at the range of the test holes would have taken months at each power level. Therefore an adjustment to a lower power level was made prior to each pressurization test. This adjustment produced a nearly constant temperature during the few days of the test. Using this technique, we were able to closely control the test hole temperatures to $\pm 1^\circ\text{C}$.

An initial test was run before energizing the heater, and then two additional tests were conducted at elevated temperatures. The heater power was then increased to full power to establish a maximum temperature plateau. After 200 hours at full power, the heater failed internally, and the test sequence had to be modified to evaluate permeability during the cool-down cycle. Two valid tests were conducted during this phase.

Finally, two of the test assemblies were moved to holes near the intersection of the orthogonal array (Fig. 1, maximum distance from the two heater holes) to evaluate the permeability of the rock mass in an area where no significant heating had occurred from any of the three tests.

Permeability Test Results. In general, the results of the permeability test are satisfactory. The deduced permeabilities are extremely low—all less than 1 nanodarcy (nd). The permeability consistently declines as the temperature increases and rises again during the cool-down phase. The last valid test during the heated phase produced data at the threshold of the resolution of the experimental apparatus (0.02 nd). The *in situ* test data are consistent with laboratory measurements made on other granitic rocks and confirm that although the Climax granite is fractured, it is an extremely impermeable medium.

Demobilization. Following the permeability test sequence, the experimental apparatus was disassembled and removed from the test area. The various transducers were returned to Livermore and recalibrated to confirm their post-test condition. The back-up data acquisition system and the cabling connecting the surface recording facility and the test area were retained for use in support of the spent-fuel test described in Sec. 2.2.

L. Ballou

2.2 Spent-Fuel Test

There are few locations where the emplacement of high-level nuclear waste can be tested in an existing

Federally-owned facility with an active safety and environmental program. One such location is the Nevada Test Site (NTS) operated by DOE primarily for the testing of nuclear explosives.

The Climax granitic stock (Climax granite) at NTS has underground facilities originally developed in the 1960s for weapon effects tests. These facilities were refurbished during FY 1977 and 1978 for Heater Test No. 1 (Sec. 2.1). They include a vertical shaft (2 m $\times$ 4 m) to the 420-m depth level, a headframe and hoist, a ventilation system, electrical utilities, and other support facilities. Thus, the Climax granite can be readily developed for test storage of spent reactor fuel in a geometry and medium generically representative of a crystalline rock repository.

Another key facility at NTS is the Engine-Maintenance and Disassembly (E-MAD) facility, where spent-fuel shipments can be received, encapsulated in canisters suitable for geologic storage, stored on an interim basis, and transferred to an LLL-furnished transporter. There is some spent-fuel-characterization capability there, and consideration is being given to establishing a calorimetry facility.

The concept of test storage of high-level waste in the Climax granite has evolved over a period of two years. In October 1976, in response to a request from DOE headquarters (then ERDA), LLL assisted the DOE Nevada Operations Office (DOE-NV) in preparing a letter outline of a concept for storage of a single canister of simulated high-level waste (using Sr or Cs capsules). Although no further action was taken at the time, a request for an update led to a January 1978 technical concept for test storage of 15 canisters of spent fuel. Funding was received in June 1978 for initial design and evaluation, and an extensive revision of the technical concept was furnished to DOE headquarters in September of 1978. This document is in the final stages of revision, and only general details are included in the present summary. The layout planned for the spent-fuel test is shown in Fig. 4.

2.2.1 Rationale

There are a number of reasons for carrying out a field test of spent-reactor-fuel emplacement. It is not widely understood by the public that spent-fuel handling is a straightforward engineering problem, involving off-the-shelf technology. For this reason, such a test would have great educational value. In addition, there are technical benefits to be gained. Unless one carries out generic tests such as this, the first test emplacement of high-level waste will not occur until we are ready to build an actual repository,

FIG. 4. Layout of 420-m level, Climax granite, showing new construction for spent-fuel test.

and this is a decade or more away. Also, even with laboratory measurements and computer modeling, we can only address the question of negative synergistic effects during storage in an actual field test. Thus the technical purpose of the test is to provide early field measurements of the effects of high-level-waste emplacement in a crystalline igneous rock.

2.2.2 Technical Issues

The spent-fuel test in Climax granite will provide:

- A simulation of the early-time thermal history of a typical waste repository element.
- A comparison of the effects of heat alone versus heat plus radiation.

The hypothetical waste repository is a large array of parallel drifts spaced on 15-m centers, with canisters 3 m apart. The spent-fuel-test simulation is a 15 m X 15 m module of that repository array (Fig. 5). The basic design comparison parameter is the calculated temperature history of the rock wall adjacent to the center canisters of both the hypothetical repository and the simulation module (Fig. 6). Calculated temperatures within the simulation module match the repository calculation within 1% in the center and 10% at the corners. Instrumentation will be deployed to measure temperature and displacement within the module and elsewhere in the array. Measured temperatures and displacements will be compared with predictions from the test design calculations.

One postulated negative effect of radiation and heat together is degradation of the heat dissipation ability of the rock. We will compare the effects of heat and radiation by using both spent fuel (a source of both heat and radiation) and electrical simulators (a source of heat). As shown in Fig. 5 there will be 11 spent-fuel assemblies and 6 chemical simulators emplaced in drill holes in a central canister-storage drift so that a statistical evaluation of the thermal response of the rock can be made. Pre- and post-test physical characterization of the rock will reveal other effects, such as decline in strength.

A number of lesser technical issues will be addressed in the experiment:

- The magnitude of displacement effects for mining alone will be documented (for comparison with thermally-induced displacement).
- The effects of mine ventilation on heat dissipation from the waste will be evaluated quantitatively.
- Test coupons of relevant materials will be included on the outside of storage hole liners (for documentation of combined radiation heat fluid-chemistry effects).
- To the extent possible in the final "as-built" configuration, the effects of anisotropy in the rock (shear or fracture zones and local mineralogic variation) will be evaluated.
- If possible, as a midtest add-on, we will evaluate various backfill materials for heat dissipation and stability in an *in situ* heat and radiation field.

FIG. 5. Layout of spent-fuel test.

FIG. 6. The temperature/time curves for the repository calculation and the spent-fuel-test calculation agree within 1% for the first 7.5 years.

2.2.3 Handling Operations

The minimum sequence of operations involving the spent fuel is as follows:

- Acquire fuel assemblies.
- Ship fuel to NTS.
- Characterize fuel pre-test.
- Encapsulate fuel in canisters at E-MAD.
- Transport to Climax granite site.
- Lower to 420-m level through dedicated access hole.
- Transfer via railcar to storage hole.
- Emplace in steel-lined storage holes.
- Energize auxiliary heaters for repository simulation.
- Monitor thermal history of fuel, electrical simulators, liners, and rock.
- Monitor radiation and rock mechanics measurements.
- Store for up to five years.
- Retrieve and return to E-MAD for post-test characterization.

In addition, more thorough, destructive characterization of one or more fuel assemblies may be carried out as an adjunct to this test. During the test, fuel-switching operations may be carried out for calorimetry, and midtest changes may include the addition of tests involving backfill in the annulus between the liners and the rock.

2.2.4 Activities in FY 1978

In addition to planning and conceptual design activities, construction and detailed design activities were initiated in FY 1978. At the close of the fiscal year, the surface preparation for mining and drilling activities was essentially complete, and work had started on refurbishing the shaft to support the heavy construction required for the underground facilities. Lag storage was under construction at E-MAD. Criteria for drilling the access hole and for underground mining had been submitted.

L. Ramspott

2.3 Planning For Rock-Mechanics Test

Heater Test No. 1 was planned and executed in a relatively short time because the technical issues were straightforward. And indeed, the test showed that excellent prediction of the thermal field in the near-field part of a granite repository is possible with available mathematical models (Sec. 2.1). An over-test in terms of thermal flux and gradient had no deleterious effects on the ability of the rock to dissipate heat under the conditions tested.

Although near-field thermal predictions seem well in hand, we must address a number of other

issues to confidently design a repository in crystalline igneous rock:

- Despite the fact that thermal overloads did not cause deterioration of a 48-mm-diam hole, a 305- to 460-mm-diam hole may respond differently. A full-scale test is needed.

- Preliminary design calculations for the spent-fuel test (Sec. 2.2) showed that expected displacements for thermal loads from spent fuel or older reprocessed waste are near the sensitivity limits of available displacement-measuring devices. Validation of mathematical models is therefore not likely with the test layout planned for the spent-fuel test.

- Currently available field instrumentation is not designed for stability through years of time in a changing thermal field, nor is it designed to measure very small effects.

- An engineering overtest is desirable; in the extreme, this would be a test to failure.

During the last half of FY 1978, work on a technical concept for a Rock-Mechanics Test Facility led to a plan to address the issues outlined above. The experimental program would be divided into three tasks, the first two of which should be carried out simultaneously:

- Overdriven field experiments
- Instrument development
- Realistic field experiments

The overdriven field experiments will produce motions significantly larger than those expected from a realistic repository situation. Thus, we will be able to measure them with reasonable accuracy using currently available measurement devices. These experiments will also constitute an engineering overtest to establish a margin of safety and to allow a preliminary evaluation of code capabilities. Tests to failure will be included if feasible.

While the overdriven field tests are in progress, an instrument-development program will be carried out. The purpose of this program will be to increase the measurement sensitivity, thermal insensitivity, and temporal stability of the measuring devices. In general, an order of magnitude improvement in measurement sensitivity would be desirable.

Finally, the improved instrumentation and codes will be used in field tests with realistic repository thermal loads to verify the codes' predictive capabilities directly, without extrapolation of non-linear responses. This phase of the test would also include a strong effort to tie field and laboratory measurements together. Thus when a site is preliminarily identified, laboratory measurements could be used in predictive codes for repository design calculations.

L. Ramspott

3. LABORATORY ROCK MECHANICS MEASUREMENTS

3.1 Physical/Chemical Factors — Water Transport In Deep Silicate Rocks

The overall goal of this study, which began in June 1976, is to assess the suitability of rocks other than salt to serve as safe, long-term containment materials for high-level radioactive wastes. We have three specific objectives leading to this goal:

- To develop working hypotheses and a theoretical basis to account for the very low permeabilities of deeply buried rocks.
- To test these hypotheses in the laboratory and in the field.
- To synthesize the laboratory and field test data and develop a predictive capability for long-term containment.

Our study focuses on igneous intrusive rocks, or high-grade crystalline metamorphic rocks.

3.1.1 Scope of Work

To predict the long-term behavior of certain igneous and metamorphic rocks, we will measure the physical properties of samples in the laboratory at simulated, deep *in situ* conditions and then correlate the data with field observations. This will allow us to predict (on the basis of minimal field exploratory data) the response in space and time of a mined cavity in these rocks.

Specifically, we are measuring the permeability, acoustic velocity, electrical conductivity, principal stresses, and principal strains of fractured and unfractured rocks loaded over a range of hydrostatic pressure and deviatoric stress. The detailed field data that we will require for correlation with laboratory measurements include fracture intensity and spacing (at the surface and at depth) and as many of the above properties as are available. We hope to develop simple models for estimating the permeability and mechanical stability of the rocks based on the mechanisms of fracture closure that we identify from the combined laboratory and field results.

3.1.2 Activities Prior to FY 1978

Prior to FY 1978, we selected six rocks for study on the basis of available (or planned) field and physical property measurements: Westerly granite (Rhode Island), Montello granite (Wisconsin), Stripa granite (Sweden), Pile Driver granodiorite (Nevada), White Lake gneissic granite (White Lake, Ontario), and Creighton gabbro (Sudbury, Ontario). Four of these materials were on hand and had been

prepared for testing in the unfractured condition. The Stripa granite and Pile Driver granodiorite were scheduled for delivery in the first quarter of FY 1979.

A high-pressure testing apparatus was constructed to measure permeability (k), acoustic velocity (V_p), and electrical conductivity (ρ) as functions of the three principal stresses (σ) and strains (ϵ). This unit was partially debugged and about one-half calibrated at the end of FY 1977. All electronic systems were complete for use in the manual mode only. Electronic components to provide automatic data acquisition for k , σ , and ϵ were complete; those for V_p and ρ were on order. The programmable capability for controlling the total experiment (and the automated data acquisition system) was completed but not debugged.

During this initial shake-down phase, preliminary measurements of V_p and ρ were completed on the Westerly granite and White Lake gneissic granite. Three reports were issued during this period. Two dealt with certain experimental aspects of the project, and the third predicted the time- and temperature-dependent closure of salt *in situ*.

3.1.3 Activities In FY 1978

Control of the experiment was completely automated through the use of an LSI-11 micro-computer. In addition, the data acquisition for k , σ , and ϵ was automated. Extension of this capability to the ρ and V_p measurements was nearly completed in FY 1978.

The ranges of pressure, pore pressure, and stress were chosen to span the range of probable conditions that might exist in an acceptable repository at depths to 2000 m. Lithostatic pressures ranged up to 50 MPa, and for all measurements, pore pressures were maintained at 0.5 times the confining (lithostatic) pressure. Deviatoric stress ranged up to 0.5 times the failure stress at that pressure. Test samples were cylinders up to 15 cm in diameter and 28 cm long. Two types of sample geometries were used: (1) material selected to be as representative as possible of the sound country rock without any large-scale fractures or faults, and (2) rock samples identical to the above except that they contained an induced through-going fracture.

Our results demonstrate that very wide ranges of permeability exist from rock to rock. Further, there are large differences in the sensitivity of permeability to stress or pressure. As an example, the unfractured White Lake gneissic granite yields

permeabilities of 10^{-18} to 10^{-19} cm² (10^{-16} to 10^{-17} darcys), which appear to be unaffected by either confining pressure or deviatoric stress. The Westerly granite yields permeabilities of 4×10^{-16} cm², which decrease by a factor of two with pressure but vary by a factor of two with stress. When compared to the initial value, compressional velocities increase by 4% and conductivity decreases by 50% as pressure is increased to 50 MPa in the gneissic granite. In Westerly granite, these become 3% and 58%, respectively. At pressure, loading of this granite to 0.5 times the failure stress increases conductivity by about 20%. Thus, it appears that V_p and ρ may be very useful tools in inferring permeability at a potential repository site or in monitoring an operational repository for permeability changes with time.

The extremely low permeabilities of the rocks we have tested so far require testing of a single sample for up to three months. Thus, our productivity has not been great, nor have we been able to make all of the tests for reproducibility that we would like. Therefore, we elected to construct a new, nearly duplicate apparatus. This new unit will accept the same large samples, will be able to make all of the measurements listed above, and will also be computer-controlled. For reasons of cost and ease of operation, we have restricted the range of permeability measurement from the original 10^{-17} – 10^{-19} cm² to 10^{-12} – 10^{-19} cm². This new unit will share some of the existing electronics with the initial apparatus. We have incorporated many new improvements into the new unit based on our experience with the first model. This new apparatus should come on-line in the second quarter of FY 1979.

H. Heard

3.2 Thermal Properties of Rocks at Elevated Temperature and Pressure

The goal of this experiment is to determine the thermal conductivity, thermal diffusivity, and thermal expansion of candidate waste repository rocks at *in situ* conditions of temperature, pressure, and fluid content. The experimental apparatus (Figs. 7 and 8) is designed for testing core samples 13 cm in diameter and 23 cm long to a range of experimental conditions which brackets likely *in situ* conditions. Temperatures will be as high as 770 K and confining pressures as high as 200 MPa. Heaters placed down a 3-cm hole along the axis of the sample are controlled such that heat flow in the central section of the sample is purely radial away from the cylinder axis. One-dimensional, axisymmetric heat-flow equations thus apply.

Thermal expansion is measured along the length of the sample.

The measurements of the three thermal properties are made in the following manner:

Thermal expansion (α). The external heaters H1, H2, and H3 (Fig. 8) are used to bring the sample to the desired temperature. The internal heaters H4, H5, and H6 are left off, and the pressure is maintained constant at the desired level. When the system reaches equilibrium (after several hours), the change in voltage in the displacement transducer from its reading at the ambient sample temperature is directly translatable to the change in length of the sample.

Thermal conductivity (k). An equilibrium temperature gradient across the sample (from its inner radius to its outer radius) is maintained by operating the internal heaters in conjunction with the external heaters. The power to the internal heaters is balanced in such a way that all the heat generated by the central section H4 flows radially. Thermal conductivity k is then calculated from the solution to the heat-flow equation in cylindrical coordinates where the radius r is the only independent variable, whence

$$q = \frac{2\pi k}{\ell \log_e(r_2/r_1)} (T_1 - T_2),$$

where q is the input power to (and output power from) heater H4, ℓ is the length of H4, r_1 and r_2 are the inner and outer radii of the sample, respectively, and T_1 and T_2 are the temperature at the inner and outer walls of the sample, respectively.

Thermal diffusivity (κ). Once the system has reached equilibrium at constant temperature and pressure, heaters H4, H5, and H6 are turned on, and the temperature pulse moving across the sample is measured with eight thermocouples at varying known radii within the sample (four are represented in Fig. 8). Again, the power to heaters H5 and H6 is slightly higher than the power to H4 so as to maintain perfectly radial heat flow in the central section where the thermocouples are located. The experiment is terminated when the outermost thermocouple indicates a temperature change of 0.1 K, estimated to be one to two minutes after the internal heaters are powered up. Assuming that the situation is symmetrical (as described above) the heat pulse obeys the relation,

$$\frac{dT}{dt} = \kappa \left(\frac{1}{r} \frac{dT}{dr} + \frac{d^2T}{dr^2} \right),$$

where r is the radial distance, T the temperature, and t time. The parameter κ is calculated by an iterative technique using repeated least-squares

FIG. 7. View of the experimental apparatus plus some of the electronics (August 1978).

Sample size: 13 cm dia X 23 cm long
 Operating limits: 773 K, 200 MPa

H1, H2, H3 — external heaters
 H4, H5, H6 — internal heaters

FIG. 8. Schematic of the experiment (with connections made).

fitting of a calculated $T(r,t)$ to the $T(r,t)$ measured by the thermocouples.

A fourth quantity, heat capacity at constant pressure C_p , is a simple function of the thermal conductivity, thermal diffusivity, and density ρ of the sample:

$$C_p = \frac{k}{\kappa \rho}$$

Progress during FY 1978 was primarily in the design, fabrication, assembly, and debugging of the apparatus. The state of the apparatus as of September 30, 1978, was as follows: a dummy sample of aluminum (unjacketed) had been run to 573 K and 63 MPa to calibrate the expansion-measurement device. The transducer used had a pressure dependence because a potted rather than an unpotted transformer winding was inadvertently used; thus, no useful data could be taken. The test

also revealed that the scheme of pressure control needed some additional valving. The bulk of the apparatus checked out positively:

- The hot and cold pressure vessels with about 50 signal and power leads remain tight at pressure and temperature.
- The external heaters deliver heat to the vessel efficiently.
- At 573 K they consumed modest amounts of power, did not heat up the cold vessel more than a couple of degrees at equilibrium, and allowed easy control of the temperature of the hot vessel.
- Data from many thermocouples (up to twelve were tested; the design is for thirty) can be read within a few seconds (roughly 0.25 sec per thermocouple) at regular, programmable intervals.

The first rock to be tested is a large-grained salt from Avery Island, Louisiana (Fig. 9). The first

FIG. 9. Sample of Avery Island salt to be tested in the apparatus. The sample, with 1/2-in.-thick ceramic end caps, is placed in an Ni-plated-Cu jacket. It has the final configuration shown at the right.

sample was machined and prepared for assembly at the close of FY 1978, but it had not yet been jacketed. The rather complex sample-loading procedure had been checked out with the run on the aluminum sample.

W. Durham

3.3 Permeability of Eleana Argillite at High Pressure

The goal of this project was to provide input to assess the suitability of the argillite in the Syncline Ridge block of the Eleana formation, NTS, for serving as a repository for permanent isolation of high-level radioactive wastes. We have studied (1) the permeability of Eleana argillite (both intact and fractured) as a function of effective pressure up to 24 MPa (equivalent to a depth of about 2 km), and (2) the effect of specimen orientation (either parallel or perpendicular to the borehole axis) on the permeability.

In FY 1978, we completed the planned permeability experiments for the Eleana argillite. We have studied seven specimens from 102, 358, and 361 m deep. All of these belong to the high-quartz group of the argillite. We have tried to prepare the low-quartz argillite specimen by coring with either water or air, but the material is too weak to be adequately prepared by either method.

Figure 10 illustrates one typical result. Our results can be summarized as follows:

- Permeability of the high-quartz argillite varies with a negative power of effective pressure.
- The intact high-quartz argillite has very low permeability, from 10^{-10} to 10^{-16} cm², depending on the effective pressure.
- One through-going fracture increases permeability by three to four orders of magnitude at low pressure and one to two orders of magnitude at a pressure of 24 MPa. At high pressure, the equivalent permeability of the fractured specimen is still very low.
- The specimens from 358 and 361 m deep have lower permeability than that from 102 m deep.
- The permeability of the tested specimens of Eleana argillite seems to be isotropic with respect to the borehole axis. This is probably due to the fact that bedding planes dip 30 to 80° with respect to the axis of the borehole.
- The effect of temperature, time, and bedding plane orientation on permeability should be thoroughly investigated.

W. Lin

FIG. 10. Permeability of Eleana argillite specimens (from a depth of 361 m, cored parallel to the borehole axis) as a function of effective pressure. The straight lines are fitted by the least-squares method.

4. LABORATORY STUDIES OF SORPTION AND LEACHING

Most of the sorption and leaching studies being conducted at LLL are supported by the Waste Isolation Safety Assessment Program (WISAP) administered by Battelle Pacific Northwest Laboratories (PNL). Other studies have been supported by DOE's Nevada Operations Office (NV). The principal goals of the WISAP program are (1) to develop generic methods to assess the safety of waste repositories in geologic media, (2) to obtain generic and site-specific data in order to apply the methods, and (3) to demonstrate assessment methods for a specific site. The requisite tasks are enumerated in Fig. 11. LLL's leaching studies in FY 1978 have been conducted in support of Task 2 and our static and dynamic sorption studies and geochemical code development work have been done in support of Task 4.

4.1 Leaching Studies of Simulated Reactor Waste Glass

An important goal of the WISAP program is the determination of the rate of radionuclide transport in the ground-water regime surrounding waste repositories. Leaching studies involving simulated and actual high-level waste glasses are being conducted to determine the magnitude of the source terms to be used in transport modeling studies.

During FY 1978, LLL has cooperated with Battelle PNL in experiments designed to compare the leach rate of simulated high-level waste glass using two markedly different leaching methods. Battelle has used the standard International Atomic Energy Agency (IAEA) leaching method, which involves sampling closed vessels periodically (Fig. 12). LLL has used a new single-pass leaching system in which fresh leachant solution is pumped over the sample at a controlled flow rate and temperature (Fig. 13). Our 420-day leaching experiments began in April 1978 and will conclude in June 1979. After completion of the studies with the simulated waste glass, we will compare the results with leaching data on actual high-level waste glass. The single-pass leaching system used in this study⁸ was developed at LLL, and it represents a modified and up-dated version of the apparatus used for leaching NTS nuclear melt glass.⁹

The simulated reactor waste used in the experiments is a sodium zinc borosilicate glass fabricated

at Battelle in the form of ellipsoidal pellets approximately 8 mm in diameter. The glass was doped with ²³⁹Pu and ²³⁷Np in concentrations that are typical of actual nuclear waste products. Nonradioactive mock fission products and ²³⁸U are also included in the glass to simulate the chemistry of actual waste.

The leachant solutions used in both experiments are distilled H₂O, a 0.03 M NaHCO₃ solution, and a synthetic brine with a composition similar to that of subsurface brines found at the Waste Isolation Pilot Plant (WIPP) Facility in New Mexico. The bicarbonate solution simulates ground water from a carbonate aquifer.

The LLL study is based on a statistically designed factorial experiment with unequal replication. Flow rate, temperature, leachant composition, and time are the independent variables; leaching rate is the dependent variable. A total of 28 simultaneous experiments are in progress: 24 on doped samples, 3 on undoped samples, and 1 on a doped sample that does not have 0.1-μm filters in the sample holder (all the others do). Volume flow rates in the single-pass apparatus are 300 ml/d, 43 ml/d, and 10 ml/d (corresponding to F, M, and S, respectively, in Fig. 14). These rates correspond to the daily, weekly, and monthly sampling frequencies in the IAEA method.¹⁰ Samples are taken at 1, 2, 3, 6, 11, 20, 37, 68, 120, 230, and 420 days. Sample temperatures are 25°C and 75°C. The modified IAEA tests at PNL are conducted at 25°C. All the PNL tests are done in triplicate. The IAEA procedure involves changing solution on the schedule described by Hesse,¹⁰ with monthly sampling continuing indefinitely. The IAEA leaching apparatus has been described by Bradley.¹¹

Preliminary results of the comparison of the two leaching methods for ²³⁷Np and the bicarbonate solution are presented in Fig. 14. The leach rate is shown as a function of time on a log-log scale. Each solid and dashed line is traced through three data points. Notable features of the plot are (1) the close agreement between the LLL and PNL data at 25°C, (2) the marked effect of temperature on the leach rate, and (3) the effect of the flow rate on leaching at 75°C.

A new single-pass leaching system is currently being designed that will incorporate rock material along with the simulated waste glass in a leach column. This second-generation experiment will commence in December 1979.

FIG. 11. Interrelationships of tasks in the WISAP program. The leaching and sorption studies being conducted at LLL are in support of tasks 2 and 4.

FIG. 12. IAEA leach-test container.

4.2 Sorption Studies

Ionic species, radioactive or stable, are selectively removed from solution during the flow of ground water through permeable rocks via ion exchange reactions between the mineral surfaces and the aqueous solution. The factors that effect the efficiency of an ion exchange reaction are ionic concentration, the nature and concentration of competing ions, the chemical nature and surface area of the solid exchange medium, contact time, and the temperature and pH of the solution.

A large volume of static or batch ionic distribution data (K_d) has been reported in the technical literature during the past three decades. There are obvious inconsistencies in methodology, which, in turn, result in wide variances in K_d values for comparable rock-water systems. One of the important WISAP goals is to evaluate the experimental methods of K_d determinations and to establish a standardized procedure for static measurements.

During FY 1978, two types of sorption studies were conducted at LLL in support of Task 4 of the WISAP program. First, an extensive series of sorption studies were carried out in the static mode

in support of a controlled-sample program in which the methodology of sorption experiments was assessed. Second, a rather sophisticated device to measure sorption in a "dynamic" mode was designed, built, and tested. Dynamic measurements of ionic distribution coefficients will eventually be compared with static measurements.

4.2.1 Static Sorption Studies in Support of the Inter-Laboratory Controlled-Sample Program

As part of the WISAP controlled-sample program, two rock types, oolitic limestone and basalt, were crushed, homogenized, characterized, packaged, and shipped to each of nine participating laboratories, where three nuclides (^{86}Sr , ^{137}Cs , and ^{237}Pu) were used in K_d determinations. Ground-water compositions were provided by PNL. The objective of the two-phase program were as follows:

- *Phase One.* To compare the experimental K_d results of the nine different laboratories using common rocks and solutions and a dictated experimental design.
- *Phase Two.* To ascertain the comparability of K_d results obtained by nine laboratories using both common and individual rocks and solutions with each laboratory using their own experimental procedure.

The experimental procedure used by LLL in the controlled sample program¹² is outlined in schematic form in Fig. 15. A synopsis of K_d results and solution compositions for all rocks tested is shown in Tables 1 and 2. The K_d values listed in Table 1 can be categorized according to the following inequalities:

- $K_{\text{Fe}} > K_{\text{Ce}}$ (except for biotite)
- $K_{\text{Ce}} > K_{\text{Sr}}$ (except for limestone)
- $K_{\text{Sr}} > K_{\text{Tc}}$

The K_{Tc} values indicate that Tc is not sorbed under the experimental conditions used in this study.

Because of LLL's long and continuing involvement with radionuclide migration studies at NTS, all of the rocks selected for the second phase of the controlled-sample program were from NTS or were in some way related to NTS lithologies. The samples from three separate shear zones in the Climax stock at NTS were selected because of LLL's current waste-isolation projects in the stock and because of future plans to conduct *in situ* tracer studies in fractured crystalline rock. The biotite sample was tested because biotite is an important accessory mineral in the Climax quartz monzonite with respect to nuclide sorption. The Thirsty Canyon tuff was selected for testing because it was the core material used in some early dynamic sorption experiments¹³ and is the initial rock to

FIG. 13. Schematic of the single-pass leaching system for simulated reactor waste glass.

FIG. 14. Results of the first three days of the LLL-PNL comparative leaching experiments. This data is for the ^{237}Np leach rate in 0.03 M NaHCO_3 solution.

FIG. 15. Batch K_a procedure for WISAP controlled-sample program.

TABLE 1. Batch K_d results for the WISAP program.

Geologic material	Ground water	K_{TC} , ml/g	K_{ST} , ml/g	K_{CS} , ml/g	K_{PU} , ml/g
Phase I					
Oolitic limestone	"Limestone"	ND	2.7 ± 18.0%	60.0 ± 50.0%	770.0 ± 4.7%
Oolitic limestone	WIPP brine B	ND	0.88 ± 49.0%	0.57 ± 94.0%	450.0 ± 20.0%
Sentinel Gap basalt	"Basalt"	ND	45.0 ± 3.0%	290.0 ± 24.0%	580.0 ± 2.1%
Sentinel Gap basalt	WIPP brine B	ND	0.0	1.6 ± 4.0%	1260.0 ± 40.0%
Phase II					
Conasauga shale	"Limestone"	0.1 ± 191.0%	29.3 ± 1.8%	4430.0 ± 0.7% 3900.0 ± 22.0%	ND
Green Argillaceous shale	"Limestone"	-0.33 ± 27.0%	60.0 ± 2.6%	8000.0 ± 25.0%	ND
Climax stock quartz monzonite (NTS)	"Granite"	-0.8 ± 26.0%	17.0 ± 7.3%	306.0 ± 0.8% 380.0 ± 35.0%	560.0 ± 27.0%
Westerly granite (Rhode Island)	"Granite"	-0.91 ± 9.1%	6.5 ± 30.0%	210.0 ± 41.0%	520.0 ± 16.0%
Biotite (Wards)	"Granite"	-1.07 ± 3.2%	59.0 ± 2.8%	663.0 ± 0.2% 680.0 ± 5.0%	570.0 ± 2.6%
Climax stock shear zone 1 (medium) ^a	"Granite"	-0.49 ± 14.0%	175.0 ± 2.0%	460.0 ± 4.6%	ND
Climax stock shear zone 2 (fine)	"Granite"	-0.6 ± 23.0%	600.0 ± 1.2%	1010.0 ± 0.5%	ND
Climax stock shear zone 2 (medium)	"Granite"	-0.7 ± 22.0%	608.0 ± 1.6%	1027.0 ± 1.5%	ND
Climax stock shear zone 2 (coarse)	"Granite"	-0.5 ± 21.0%	601.0 ± 3.8%	1061.0 ± 0.6% 1500.0 ± 47.0%	ND
Climax stock shear zone 3 (fine)	"Granite"	-0.2 ± 60.0%	440.0 ± 2.7%	1400.0 ± 1.2%	ND
Climax stock shear zone 3 (medium)	"Granite"	-0.2 ± 137.0%	311.0 ± 2.8%	940.0 ± 2.8%	ND
Climax stock shear zone 3 (coarse)	"Granite"	-0.4 ± 30.0%	318.0 ± 2.2%	1100.0 ± 5.7%	ND

^aDesignations refer to particle size of sample subsequent to crushing and sieving; fine = <325 mesh, medium = 100-325 mesh, coarse = 50-100 mesh.

TABLE 2. Chemical compositions of simulated ground water used in the WISAP controlled-sample program.

"Basalt" ground water		"Granite" ground water		"Limestone" ground water		WIPP brine (B)	
Chemical species	mg/l	Chemical species	mg/l	Chemical species	mg/l	Chemical species	mg/l
Na ⁺	30.0	SiO ₂	26.2	Ca ⁺	50.0	Na ⁺	115,000.0
K ⁺	9.0	Ca ⁺²	8.3	Mg ⁺²	3.6	K ⁺	15.0
Ca ⁺²	6.5	Mg ⁺²	1.5	Na ⁺	2.3	Mg ⁺²	10.0
Fe ⁺²	1.0	Na ⁺	6.7	K ⁺	0.4	Ca ⁺²	900.0
NO ₃ ⁻	58.0	K ⁺	1.5	HCO ₃ ⁻	153.0	Sr ⁺²	1.5
SO ₄ ⁻²	23.0	HCO ₃ ⁻	47.7	SO ₄ ⁻²	14.4	Cs ⁺	1.0
Cl ⁻	16.0	SO ₄ ⁻²	2.7	Cl ⁻	3.5	Cl ⁻	175,000.0
I ⁻	0.7	Cl ⁻	2.1			SO ₄ ⁻²	3,500.0
						BO ₃ ⁻³	10.0
						HCO ₃ ⁻	10.0
						Br ⁻	400.0
pH 7.7-8.2		pH 7.8 ± 0.3		pH 8.2 ± 0.2		pH 6.5 ± 0.5	

be evaluated in the current dynamic sorption experiments. The Bonanza King dolomite is the aquifer material at the U.S.G.S. Amargosa Tracer Site located south of NTS. As a result of K_d measurements on the dolomite, it has been ascertained that in tracer studies with Sr at the Amargosa site, nuclide transit from injection to observation wells would occur in reasonable time periods, i.e., six months or less. K_d measurements with Cs and Pu suggest that transit times for these nuclides would be unreasonably long ($\geq$ five years). The Conasauga shale and the green argillaceous shale samples were tested by all participating laboratories.

4.2.2 Dynamic Sorption Studies

An experimental apparatus (Fig. 16) was designed, built, and tested in FY 1978 for the purpose of measuring the distribution of ionic species between an exchange medium and an aqueous solution under dynamic or flow conditions. Corresponding static sorption experiments have been and will be conducted to provide a basis for evaluating the dynamic scheme. The dynamic method of determining K_d values of rock material is considered to be intermediate in character between batch K_d determina-

tions and K_d determined by means of *in situ* tracer studies. The principal objectives of the experimental plan are:

- To compare dynamic K_d determinations on cores with static K_d measurements on powdered rock.
- To study dispersion and desorption phenomena by injecting appropriate tracer pulses into a steady-state flow through a core.
- To systematically investigate sorption as a function of time, confining pressure, temperature, competing ions, ionic concentration, and flow rate.

The apparatus is designed for fluid inlet pressures of up to 3.4 MPa (500 psi) and radial confining pressures up to 34.8 MPa (6000 psi). Sample holders have been designed for samples 13 or 26 mm in diameter and 26 mm in length. The cores are covered by cast plastic jackets, which seal the core and end-fittings from invasion of confining oil. Two separate core-testing devices have been built, one of which is equipped with a high temperature bath for the core vessel (max temp = 80°C).

The initial experimental runs have been conducted with distilled water and cores of the Thirsty Canyon tuff from NTS. Radionuclide spikes have been ^{99m}Tc and ⁸⁶Sr. A typical effluent curve

for ^{137}Cs is shown in Fig. 17. In future experiments, Thirsty Canyon tuff and Climax stock monzonite will be used with distilled water, bicarbonate ground water, and "granite" ground water, whose composi-

tion is included in Table 2. Analysis of the effluent is done with a well-type gamma-ray scaler.

J. Lewhey

FIG. 16. Schematic view of the dynamic sorption apparatus.

FIG. 17. Effluent data for spike injection of Tc in distilled water with Thirsty Canyon tuff.

5. SUPPORTING R&D

Supporting R&D has been necessary in carrying out the work summarized in previous sections. For example, the development of instrumentation for the heater tests (Sec. 2.1) was a significant part of the test. In all projects, we have modified or developed computer codes to analyze the data, and in some cases we have developed codes to control the experiment.

In this section, three areas of supporting R&D have been singled out for discussion. In all three cases, waste-isolation funds constituted only a small part of the support. However, the R&D results seem to offer significant promise for application to waste-isolation problems.

5.1 Thermal Expansion Data Resulting From Laboratory Measurements of Seismic Q

The ILL Seismic Monitoring Program has funded a development effort to measure internal friction (seismic Q) in rocks at appropriate conditions of pressure and temperature. A small amount of supplemental funding was provided by the waste-isolation project. A major technical goal is to develop techniques for measuring strains at pressure and at temperatures to ~ 1200 K. When developed, this technology will provide data of value to the nuclear-waste-isolation projects. In particular, we will be able

to determine the thermal expansion of rocks at temperatures of at least 600 K under pressure sufficient to suppress crack effects. Once the "intrinsic" thermal expansion is known, the effect of micro-cracking on thermal expansion can be evaluated.

The apparatus used in preliminary tests is an internally heated gas pressure vessel capable of pressures to 1.0 GPa and temperatures to 1250 K. Strains for thermal expansion calculations are measured with a high-temperature strain-gauge system developed at Battelle Columbus Laboratory by M. M. Lemcoe and colleagues. We have been engaged in a year-long joint research effort aimed at extending this technology to strain measurements on rocks at high pressure.

During FY 1978, we have obtained promising results. With the proper surface treatment, the gauge system survives intact to ~ 1200 K in the high pressure environment. Preliminary apparent strain data have been obtained for one standard material, OFHC copper. The standard data will be used to evaluate the effect of pressure on the gauge factor for the gauge system and to make relative measurements of thermal expansion for unknowns (i.e., granitic rocks). The first measurements on Climax stock granodiorite are scheduled for early FY 1979. We expect that thermal expansion to ~ 600 K at 200 MPa will be available by Mid-FY 1979.

B. Bonner

5.2 Use of Electrical Methods in the Laboratory and Field

Electrical measurements were among the earliest applied geophysical techniques in petroleum exploration and are widely used in the mineral industries. Advances at LLL have come in two main areas: (1) the use of advanced instrumentation and (2) the application of advanced computer techniques for data reduction. The range and sensitivity of cross-borehole measurements have thereby been extended beyond previous capabilities. These applications have been reported earlier.^{14, 28} The following brief summary deals only with applications to waste isolation.

5.2.1 Monitoring the Electrical Conductivity of Core Samples

During FY 1977, a new method was devised for determining the electrical conductivity of core samples.²⁹ It was used successfully during FY 1978 to obtain the electrical conductivity data described in Sec. 3.1. This new method is especially helpful in that electrodes are placed on the sides, rather than the ends of the core sample. This permits an experimenter to monitor the conductivity as fluids are moved from the ends of the core sample through the rock. By using a number of electrode locations, the anisotropy of the rock can be measured, even in a pressure vessel and even with fluid moving through the rock. Also the location of the fluid front versus time can be determined.

5.2.2 Assessing the Detailed Interior Structure of Core Samples Using Low Frequency Probing

An outgrowth of the system described in Sec. 5.2.1 has been the idea of an "impedance camera," consisting of numerous electrodes on the sides of a core sample. By sequentially sampling varied voltages applied to these electrodes and the resultant induced low frequency currents, one can obtain a large volume of data characterizing the three-dimensional interior of the core sample. A method of analyzing these data to provide a 3-D image has been devised and is currently being tested. An experimental apparatus for generating and sampling the required number of voltages and currents is near completion and will be tested in FY 1979. This data-collection data-analysis image-presentation procedure is expected to require less than five minutes per core sample. With such a system, detailed interrogations of core samples under varied environmental conditions are possible. The initial system nearing completion uses low frequency electromagnetic probing. Later versions will use micro-

wave, ultrasonic, X-ray, γ -ray, and infrared probing to characterize the interior of the core sample. The general philosophy of the approach and examples of how it can be used in the laboratory (on core samples) and in the field (cross-borehole probing) are given in Ref. 30.

5.2.3 Use of High Frequency Electromagnetic Probing at Field Test Sites in Granitic Rocks

During FY 1978, we have been preparing for the field application of high frequency electromagnetic (HFEM) probing at the experimental sites at Stripa, Sweden (I.B1.) and NTS (I.I.1.). The field tests will be carried out during FY 1979.

In these efforts, we will be using the technique of geotomography, which has been conceived, developed, and extensively used by LLL.¹⁴ Geotomography is based upon the analysis of cross-borehole transmission data (both seismic and electromagnetic) by techniques that are analogous to those used in interpreting X-ray scans of the human body (i.e., X-ray tomography).

The purpose of the field tests is two-fold: to document inhomogeneities in the rock (i.e., fracture or shear zones); and to establish the use of electrical methods as a tool for monitoring permeability changes in the rock, by correlating the field electrical measurements with laboratory data described in Sec. 3.1.

J. Lytle

5.3 Geochemical Equilibrium Code Development

During FY 1978, a limited amount of funding has been allocated to the development of geochemical equilibrium codes that are applicable to problems associated with nuclear waste isolation in geologic media. Equilibrium codes developed at LLL have been successfully used to predict rock-water interactions in geothermal environments,³¹ and the development of codes suitable for waste-isolation problems is an extension of this earlier work. The current focus of the development effort is a code that is being produced by Dr. Howard Levine of JAYCOR Corporation of La Jolla, California under contract to LLL. In addition, the EQ3-6 code has recently been acquired, since its author, Dr. Thomas Wolery is now a member of the Earth Sciences Division of LLL.³²

The Levine code calculates the composition of geochemical species in complex aqueous systems at thermodynamic equilibrium. It is considered to be a

state-of-the-art code in that: (1) it is faster than L.L.'s existing equilibrium codes, (2) it requires only a small memory capability, (3) it is particularly effective for multiphase systems, and (4) it is general enough to handle systems containing solids, liquids, and gases. It is suitable for predicting the geochemical species that result from the processes of cooling, evaporation, and precipitation and from the mixing of dissimilar solutions.

The Levine code finds the minimum free energy of a system by solving the nonlinear equilibrium constant equations for the reactions of that system. It differs from other codes in that it considers only one reaction at a time (univariant descent) instead of considering all reactions simultaneously (multivariant or steepest descent). The fact that the code considers only one reaction at a time is significant for several reasons: (1) it reduces storage needs, (2) it simplifies arithmetic so that full machine accuracy is available, and (3) it is faster for problems of heterogeneous equilibria. Thus, problems that might arise from large scale matrix inversion and unstable numerical solutions are eliminated. The code will also converge for problems that other existing codes cannot handle, such as the decomposition of carbonates and systems with multiple oxidation states of widely separated electrochemical potential.

Central to both the Levine and EQ3 6 codes is the calculation of the distribution of species and phases in a system for which the bulk (elemental) composition, temperature, and pressure are specified. EQ3 6 uses a highly modified Newton-Raphson technique to perform the calculation, and this capability serves as a subroutine under the control of a "driver" code to simulate irreversible reaction of components in aqueous systems. The geochemical foundation of such predictive models has been discussed by Helgeson.^{11, 12} The numerical foundation and modeling capabilities of EQ3 6 are superior to those of Helgeson's original code PATHI, for which EQ3 6 was explicitly written as a replacement. The driver function of the newer code permits simulation of simple titration processes and irreversible reaction in both closed and flow-through systems. Temperature and pressure may change with reaction progress in these models. However EQ3 6 requires an aqueous phase and has not yet been adapted to include gases.

The driver function of the Levine code is currently primitive compared to that of EQ3 6. The central equilibrium calculation function, however, is clearly superior to that of EQ3 6. Two possibilities are currently under study: incorporating the Levine code into EQ3 6, or expanding the current Levine code driver to match that of EQ3 6.

These codes are superior for predicting the geochemical species that result from processes of heating, cooling, evaporation, precipitation, and the mixing of dissimilar solutions. More specifically, the types of nuclear waste problems that can be addressed are:

- The nature of the geochemical interaction among the waste form, container material, and the coexisting aqueous phase.
- The geochemical interaction between dissolved species and the enclosing geologic media, including backfill.
- The calculation of what type of material would immobilize a particular dissolved species by means of precipitation and/or absorption.

The point should be made that these geochemical codes are based on the concept of thermodynamic equilibrium, and it is well known that thermodynamics alone does not provide information on reaction rates. In many chemical reactions, thermodynamic considerations tend to be overshadowed by kinetic factors. However, the geochemical processes associated with the subsurface disposal of waste tend to be infinitesimally slow (e.g., the movement of ground water), suggesting that the geochemical reactions occurring in such an environment may approach equilibrium. Thus, an equilibrium code would be particularly applicable to these regimes. Moreover, reactions close to equilibrium may possibly be described in terms of relative rates,¹³ still making equilibrium codes a valuable tool.

The Levine code was 90% complete as of the end of FY 1978, and it should be operational in early FY 1979. The EQ3 6 code is already complete, although improvements are still being made. Copies have been shared on request with other research agencies, including the Lawrence Berkeley Laboratory.

T. Wolery
D. Miller

6. QUALITY ASSURANCE

Quality Assurance (QA) at L.L. is guided by the *L.L. Quality Assurance Manual*.¹⁶ Personnel in the various waste-isolation projects are aware of the need for QA, and each task group is planning implementa-

tion through written QA plans and procedures. The first formal plan (for the spent-fuel test described in Sec. 2.2), was in draft form at the end of FY 1978 and was issued November 1, 1978.

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APPENDIX A. ABSTRACTS OF REPORTS ISSUED AND TECHNICAL PRESENTATIONS MADE DURING FY 1978

D. G. Coles, H. C. Weed, D. D. Jackson, and J. S. Schweiger, *Single-Pass Leaching of Nuclear Melt Glass by Groundwater, Part I*, Lawrence Livermore Laboratory, Preprint UCRL-81617, Part I (1978). Presented at American Chemical Society Symposium, Miami Beach, Florida, September 11-15, 1978.

Melt glass (fused rock containing fission and activation products and actinides) produced by three underground nuclear explosions has been leached at 25°C with groundwater obtained from a well near the explosion sites. Leaching was carried out in cells with the groundwater passing over glass particles (size range: 40-130 μ) at flow rates of 36 and 6.7 cm/day. With this dynamic flow-through system, the solid sample is continually exposed to fresh leaching solution so as to simulate actual underground conditions. Radionuclide concentrations in the leachate were obtained at intervals of 1, 2, 3, 6, 11, 32, 38, 70, and 120 days after the leaching began. The fraction of radioactivity leached per day per m^2 surface area varied from $1.5 \cdot 10^{-2}$ for ^{22}Na on Day 1 to 7.0×10^{-6} for ^{144}Ce on Day 120. Except for ^{144}Ce , leaching rates did not seem to depend significantly on flow rates.

D. G. Coles, H. C. Weed, and J. S. Schweiger, *Single-Pass Leaching of Nuclear Melt Glass by Groundwater, Part II*, Lawrence Livermore Laboratory, Preprint UCRL-81617, Part II (1978). To be presented at American Chemical Society Meeting, Honolulu, Hawaii, April 1-6, 1979.

Nuclear explosion melt glass has been continuously leached with Nevada Test Site (NTS) groundwater for 420 days. A single-pass dynamic flow system was used in order to simulate actual underground conditions. The experimental details and preliminary data for the first 120 days of leaching of only one of the three glass samples have been presented earlier (1). This discussion presents the final data for all three glass samples leached during the experiment. Leaching was carried out at flow rates of 36 and 6.7 cm/day and at 25°C. The samples ranged from 40 to 130 μm in particle size and contained from 65 to 100 percent glass. The highest leaching rate observed was for ^{22}Na and varied from $1.5 \times 10^{-2} \text{ day}^{-1} m^{-2}$ on Day 1 to $6 \times 10^{-8} \text{ day}^{-1} m^{-2}$ on Day 420. The lowest leaching rate observed was for ^{144}Ce and varied from $1.5 \times 10^{-6} \text{ day}^{-1} m^{-2}$ on Day 1 to $4.5 \times 10^{-6} \text{ day}^{-1} m^{-2}$ on Day 420. Cerium-144 showed the greatest dependence of leaching rate on flow rate. Mechanisms are presented which describe the leaching behavior of the glasses with respect to radionuclides observed in the leachate. Surface effects as observed by SEM on the glass particles after the leaching was terminated are also discussed.

H. D. Heard, D. Trimmer, W. Lin, B. Bonner, and A. Duba, *Permeability of Generic Repository Rocks at Simulated In Situ Conditions*, Lawrence Livermore Laboratory, Preprint UCRL-81767 (1978). To be presented at Symposium on Underground Disposal of Radioactive Wastes, IAEA, Otaniemi, Finland, July 1979.

The safe terminal storage of solidified high-level radioactive wastes in a mined repository in the earth's crust requires that the time constant for fluid movement from the repository to the biosphere be long compared to the decay of the waste. Further, adequate diagnostic monitoring of the repository is necessary to provide assurance that all changes are in accordance with predictions.

We report here new measurements of the permeability of large rock samples of igneous and metamorphic rocks over a range of confining pressure, pore (water) pressures, and deviatoric stress. Seven rocks are being evaluated: three granites, a granodiorite, a gabbro, a gneissic granite, and an argillite. The range of pressure, pore pressure, and stress were chosen to span the range of probable conditions which might exist in an acceptable repository at depths to 2000 m. For all measurements, pore pressures were maintained at 0.5 of the confining (lithostatic) pressure. Deviatoric stress ranged up to 0.5 of the failure stress at that pressure. Test samples were cylinders up to 15 cm in diameter by 28 cm long. Two types of sample geometries were used: (1) material selected to be as representative as possible of the sound country rock without any large-scale fractures or faults, and (2) rock samples identical to (1) except containing an induced through-going fracture.

Simultaneously we also determine the compressional wave velocity and electrical conductivity. A major purpose here is to establish a quantitative relationship among pressure, stress, strain, permeability, velocity, and conductivity through each rock at likely repository conditions.

R. J. Lytle and K. A. Dines, *An Impedance Camera: A System for Determining the Spatial Variation of Electrical Conductivity*, Lawrence Livermore Laboratory, Rept. UCRL-52413 (1978).

A data collection and data interpretation method is presented for predicting, from measurements made on the periphery of the core sample, the electrical conductivity distribution within core samples. This method uses an array of electrodes on the periphery to probe through the core sample. Surprisingly accurate detail can be seen in the estimated conductivity distribution. This method has been demonstrated using synthetic examples modeled and analyzed on a computer. Extensions of the procedure may be useful in subsurface geophysical probing and remote probing methods using physical phenomena satisfying Laplace's equation.

R. J. Lytle, A. G. Duba, and J. L. Willows, *Alternative Methods for Determining the Electrical Conductivity of Core Samples*, Lawrence Livermore Laboratory, Preprint UCRL-81695 (1978). Prepared for submittal to *Review of Scientific Instruments*.

Electrode configurations are described that can be used in measuring the electrical conductivity of a core sample and that do not require access to the core end faces. The use of these configurations eliminates the need for machining the core ends for placement of end electrodes. This is because the conductivity in the cases described is relatively insensitive to the length of the sample. We validated the measurement technique by comparing mathematical models with actual measurements that were made perpendicular and parallel to the core axis of granite samples.

S. C. MacLean, D. G. Coles, and H. C. Weed, *The Measurement of Sorption Ratios for Selected Radionuclides on Various Geologic Media*, Lawrence Livermore Laboratory, Rept. UCID-17928 (1978).

The distribution ratio R_D is measured in the same way as the sorption coefficient K_D , but differs from K_D in that R_D is not necessarily an equilibrium value. Distribution ratios (R_D) have been obtained for ^{87}Sr and ^{137}Cs on various geologic materials including limestone, basalt, dolomite, granite, tuff, and shale. R_D values for ^{237}Pu were determined on all the materials listed above, except shale. R_D values for $^{99\text{m}}\text{Tc}$ were determined on biotite, granite, tuff, and shale. The R_D values in this study obey the following inequalities:

$$\begin{aligned} R_D(\text{Pu}) &> R_D(\text{Cs}) \text{ except for biotite} \\ R_D(\text{Cs}) &> R_D(\text{Sr}) \text{ except for limestone} \\ R_D(\text{Sr}) &> R_D(\text{Tc}). \end{aligned}$$

Tc is not sorbed under the conditions of this study.

L. D. Rampott (technical editor), *Waste Isolation Projects X FY 1977*, Lawrence Livermore Laboratory, Rept. UCRL-50050-77 (1978).

(no abstract)

D. R. Stephens and W. Lin, *Relationships Among Permeability, k , Porosity, η , and Formation Factor, F* , Lawrence Livermore Laboratory, Preprint UCRL-80856 (1978).

We derive some useful relationships among permeability, porosity, and formation factor, and compare derivations with previous works, some of which are in error. The simple equation,

$$k = \frac{m^2}{k_{11} F^2}$$

is shown to be in reasonable agreement with experimental data for a number of rocks.

H. C. Weed, D. G. Coles, D. J. Bradley, R. W. Mensing, and J. S. Schweiger, *Leaching Characteristics of Actinides from Simulated Reactor Waste Glass*, Lawrence Livermore Laboratory, Preprint UCRL-81147 (1978). Presented at Symposium on Science Underlying Radioactive Waste Management, Boston, Massachusetts, November 28–December 1, 1978.

Two methods for measuring the leach rates of simulated high level waste glass are compared. One is a modification of the standard IAEA method and the other is a one-pass method in which fresh leachant solution is pumped over the sample at a controlled flow rate and temperature. For times up to 3 days, there is close agreement between results from the two methods at 25.0C. Leach rates from the one-pass method show no correlation with flow rate at 25.0C, but at 75.0C leach rate increases with flow rate. Np-237 rates at 75.0C are greater than those at 25.0C, but ^{239}Pu rates at 75.0C are less than or equal to those at 25.0C. Resorption of ^{239}Pu associated with $(\text{SiO}_2)_n$ polymers at high temperature is suggested as a possible cause.