

324  
6-10-80  
JWES

OR. 1290

**LA-8309-PR**

Progress Report

**MASTER**

**Research and Development Related to the  
Nevada Nuclear Waste Storage Investigations**

**October 1 — December 31, 1979**

*cr*  
University of California



**LOS ALAMOS SCIENTIFIC LABORATORY**

Post Office Box 1663 Los Alamos, New Mexico 87545

REPRODUCTION OR USE OF THIS DOCUMENT IS UNLIMITED

## **DISCLAIMER**

**This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.**

---

## **DISCLAIMER**

**Portions of this document may be illegible in electronic image products. Images are produced from the best available original document.**

**An Affirmative Action/Equal Opportunity Employer**

Previous reports in this series, unclassified, are  
LA-7647-PR, LA-7900-PR, LA-7974-PR, and  
LA-8152-PR.

This report was not edited by the Technical  
Information staff.

This report was prepared by the Los Alamos  
Scientific Laboratory as part of the Nevada  
Waste Storage Investigations managed by the  
Nevada Operations Office of the US Department  
of Energy. Based upon their applicability to the  
investigations, some results from the Radionuclide  
Migration Project, managed by the Nevada  
Operations Office of the US Department of  
Energy, are included in this report.

This report was prepared as an account of work sponsored  
by the United States Government. Neither the United  
States nor the United States Department of Energy, nor  
any of their employees, makes any warranty, express or  
implied, or assumes any legal liability or responsibility for  
the accuracy, completeness, or usefulness of any information,  
apparatus, product, or process disclosed, or represents  
that its use would not infringe privately owned rights.  
Reference herein to any specific commercial product,  
process, or service by trade name, mark, manufacturer, or  
otherwise, does not necessarily constitute or imply its  
endorsement, recommendation, or favoring by the United  
States Government or any agency thereof. The views and  
opinions of authors expressed herein do not necessarily  
state or reflect those of the United States Government  
or any agency thereof.

**UNITED STATES  
DEPARTMENT OF ENERGY  
CONTRACT W-7405-ENG. 36**

LA-8309-PR  
Progress Report  
UC-70  
Issued: April 1980

# **Research and Development Related to the Nevada Nuclear Waste Storage Investigations**

**October 1 — December 31, 1979**

Compiled by

**Bruce M. Crowe**

Contributors

<b>B. P. Bayhurst</b>	<b>F. O. Lawrence</b>
<b>J. Blacic</b>	<b>C. Nelson</b>
<b>B. M. Crowe</b>	<b>J. R. Smyth</b>
<b>W. R. Daniels</b>	<b>M. Sykes*</b>
<b>S. J. DeVilliers</b>	<b>J. L. Thompson**</b>
<b>B. R. Erdal</b>	<b>E. N. Vine</b>
<b>F. Goff</b>	<b>D. Vaniman</b>
	<b>K. Wolfsberg</b>

\*Short-Term Visiting Staff Member, Arizona State University, Tempe, AZ 85281.  
\*\*Short-Term Visiting Staff Member, Idaho State University, Pocatello, ID 83209.

DISCLAIMER

This book was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise, does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.



DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

ley

RESEARCH AND DEVELOPMENT RELATED TO THE  
NEVADA NUCLEAR WASTE STORAGE INVESTIGATIONS

October 1 - December 31, 1979

Compiled by

Bruce M. Crowe

ABSTRACT

Batch sorption measurements in air with strontium, cesium, barium, cerium, and europium have been started with four additional tuff samples to extend the data base of sorptive properties of Yucca Mountain tuff with type and mineralogy. Additional results for sorption of plutonium and americium are presented. Results for sorption of technetium, strontium, cesium, barium, cerium, europium, and uranium(VI) in a nitrogen atmosphere (<0.2 ppm oxygen and <20 ppm carbon dioxide) are given and compared with results in air. Migration rate studies with crushed tuff are continuing, and, in some cases, uniform "peakless" elution is being observed. Methods for studying the migration rate of uranium(VI) are being developed. Nuclides are being infiltrated into various types of solid cores for study after sectioning. Microautoradiographic examination of crushed tuff samples used in the batch determinations indicate that large species of americium are sorbed. Methods for determination of iron(II) in both rocks and waters are described, and a method for determining the redox potential of ground water is being developed. A field test of radionuclide migration at G-tunnel at the Nevada Test Site will be funded by the Office of Nuclear Waste Isolation with Los Alamos Scientific Laboratory as the lead organization.

A new Siemens automated powder diffractometer is being installed and is operational in the manual mode. X-ray diffraction patterns for core samples from UE25a-1 were rerun with the new equipment. Preparation work for initiating the hydrothermal soak tests

is nearly complete. Some difficulty has been encountered in developing a jacketing system for high-temperature runs. A new creep-testing apparatus has been designed with a computer-controlled stress actuator. Construction will begin shortly. Geologic field mapping in the Reveille and Pancake ranges is continuing. The probability of disruption of a repository site at Yucca Mountain by future volcanic activity is bracketed by the range  $10^{-8}$  to  $10^{-10}$ /yr. Partitioning functions for selected elements in plagioclase and olivine phenocrysts were examined for estimates of near-liquidus temperatures of representative melt compositions of the basalts of Crater Flat. Higher temperature estimates were calculated for glomerophyritic clots in the oldest basalts of Crater Flat. These data are consistent with either high  $\text{pH}_2\text{O}$  during crystallization of the clot phases, or the clots may be xenocrystic with respect to the host rock.

Implementation of the Quality Assurance Program was continued with development of work plans and procedures and surveillance of assigned subtasks. Work was continued on development and implementation of the U.S. Geological Survey Quality Assurance Program.

---

## I. MEDIA INVESTIGATIONS

### A. Strontium, Cesium, Barium, Cerium, and Europium

Sorption measurements of strontium, cesium, barium, cerium, and europium on a group of six Yucca Mountain tuff samples from drill hole UE25a#1 were reported previously.<sup>1</sup> Those measurements indicated that, particularly for strontium, cesium, and barium, the presence of zeolites is important for high sorption ratios. To expand those studies and, hopefully, to better relate them with measurements made earlier<sup>2</sup> on tuffs from Jackass Flats, Nevada, four additional Yucca Mountain tuffs are being studied by a batch technique in air.<sup>1</sup> For comparison, measurements with sample JA-18 are being repeated.<sup>2</sup>

Sample YM-30 is a densely welded devitrified tuff with zeolite-containing fractures. Sample YM-5 is a fresh (glassy) nonwelded tuff without zeolites. Sample YM-42 is an immature sandstone with clay matrix, and sample YM-46, a devitrified densely welded vitric crystal tuff, has heavy iron staining but no zeolites.

Only measurements for a sorption time of 3 weeks have been completed to date. Preliminary sorption ratio values are given in Table I.

#### B. Uranium(VI)

The results of the measurement of the sorption behavior of U(VI) on Yucca Mountain cores under atmospheric conditions show average sorption values of 2 to 5 me/g and desorption values of 6 to 15 me/g. As noted in the discussion of controlled atmosphere (<0.2 ppm oxygen and <20 ppm carbon dioxide) measurements of the same cores,<sup>1</sup> U(VI) is more highly sorbed on the YM-38 sample than on samples from the other cores investigated, but it is a factor of  $\sim 3$  more sorbed under the controlled nitrogen atmosphere than in air. This could be due to the difference in carbonate concentration with loss of carbon dioxide in the controlled nitrogen atmosphere.

#### C. Plutonium and Americium

Details of the batch experiments for the measurement of the sorption of plutonium and americium on Jackass Flats tuff under aerobic conditions are given in Ref. 1. Additional measurements have been made, primarily the analyses of <sup>239</sup>Pu in both solid and aqueous phases from Experiment 2 and the completion of 6-month contact times for <sup>241</sup>Am in Experiment 4. Table II updates the results given previously.<sup>3</sup>

The general conclusions for these experiments are given in Ref. 1. The additional results for americium tend to confirm the observation that sorption ratios ( $R_d$  values) for the sorption of americium on tuff show very little increase with longer contact times, at least in the range from a few weeks to 6 months. The <sup>239</sup>Pu results did nothing to clarify the confusing situation in regard to plutonium sorption. Comparison of results for  $\approx 4 \times 10^{-7}$  M <sup>239</sup>Pu and  $\approx 1 \times 10^{-13}$  M <sup>237</sup>Pu shows higher sorption  $R_d$  values for the more concentrated solutions but higher desorption  $R_d$  values subsequent to contacts with the less concentrated solutions, at least for "pH adjusted" feed solutions. Also, for the "concentrated" <sup>239</sup>Pu solutions the sorption  $R_d$  values are greater than the desorption values, but for the "dilute" <sup>237</sup>Pu solutions the desorption  $R_d$  values are greater than the sorption values. For the "dried" method of preparing the feed solutions, the desorption  $R_d$  values in all cases are greater than the sorption  $R_d$  values. These results are probably the effects of plutonium speciation and associated solubility differences in the two methods of feed solution preparation. Studies of the fundamental chemistry of

TABLE I  
SORPTION RATIOS (mL/g), THREE-WEEK CONTACT TIME

	<u>Sr</u>	<u>Cs</u>	<u>Ba</u>	<u>Ce</u>	<u>Eu</u>
YM-42	2 600	15 000	64 000	28 000	37 000
YM-5	180	3 800	600	130 000	120 000
YM-30	94	760	1 800	110 000	7 700
YM-46	81	400	5 800	80 000	93 000
JA-18	14 000	14 000	86 000	1 200	1 400

TABLE II  
AVERAGE SORPTION RATIOS (mL/g) FOR Am AND Pu<sup>a</sup>

<u>Element</u>	<u>Core</u>	<u>Temp</u> (°C)	<u>Dried Feed Preparation</u>		<u>pH-Adjusted Feed Preparation</u>	
			<u>Sorption</u>	<u>Desorption</u>	<u>Sorption</u>	<u>Desorption</u>
Am	JA-18	22	180(30)	1 100(260)	435(6)	960(15)
		70	230(30)	3 400(300)		
	JA-32	22	130(30)	2 200(650)	1 200(100)	2 300(310)
		70	110(30)			
	JA-37	22	670(210)	17 000(3 500)	11 000(1 500)	12 000(2 000)
		70	970(240)			
Pu	JA-18	22	120(20)	340(110)	1 200(210) <sup>b</sup>	750(170)
		70	110(20)			
	JA-32	22	110(20)	1 100(640)	1 200(210) <sup>b</sup>	750(170)
		70	74(14)			
	JA-37	22	500(220)	8 200(1 800)	3 300(1 200) <sup>b</sup>	3 800(950)
		70	700(240)			

<sup>a</sup>Values in parentheses are the standard deviations of the means (absolute values) for replicate measurements.

<sup>b</sup>Value appeared in wrong column in April 1-June 30, 1979 quarterly report (NVO-196-11 and LA-7974-PR).

plutonium, americium, and other actinides in near-neutral solutions are essential to understanding the sorptive behavior of these elements.

## II. SORPTION UNDER A CONTROLLED NITROGEN ATMOSPHERE

### A. Technetium

Measurements by a batch technique of the sorption ratios of  $^{95m}\text{Tc}$  on three samples of Yucca Mountain tuffs under a controlled atmosphere of nitrogen (<2.0 ppm oxygen, <20 ppm carbon dioxide) were performed. Two fraction sizes (<75 $\mu\text{m}$  and 75 to 500  $\mu\text{m}$  were studied; however, little difference in sorption ratios between the two fractions was observed. Sorption ratios increased with time (from 3 to 12 weeks). The average sorption and desorption ratios (for both fraction sizes and all times) are given in Table III. The highest  $R_d$  values were obtained for core YM-38, the only zeolite-containing tuff studied, although desorption ratios for all three tuffs were approximately ten times larger than the corresponding sorption ratios. In previous measurements with  $^{95m}\text{Tc}$  on tuffs in air, the average  $R_d$  values were 0.2  $\text{m}\ell/\text{g}$  and 1.5  $\text{m}\ell/\text{g}$  for sorption and desorption, respectively. Thus sorption ratios of technetium on tuffs are 10 to 100 times larger under the nitrogen atmosphere than in air.

### B. Strontium, Cesium, Barium, Cerium, and Europium

Batch sorption determinations with strontium, cesium, barium, cerium, and europium under the nitrogen atmosphere have been performed using tuff samples of Yucca Mountain cores 22, 38, and 54. Measurements were made for 3-, 6-, and 12-week sorption times and 3- and 6-week desorption times. The sorption ratios are listed in Table IV. All determinations were made at ambient temperature. The  $R_d$  values obtained for these elements on all the Yucca Mountain samples are consistent with the values obtained under atmospheric conditions. Only the  $R_d$  values for barium, and, in a few cases, strontium, seem to be lower (by a factor of 2) than those measured under normal atmospheric conditions. The small difference may be explained by the absence of carbon dioxide in the controlled nitrogen atmosphere. In almost all cases values from desorption experiments are higher than those from sorption experiments. This was also observed in the measurements under atmospheric conditions.

TABLE III  
SORPTION RATIOS OF  $^{95m}\text{Tc}$  ON TUFFS UNDER CONTROLLED  
ATMOSPHERIC CONDITIONS

<u>Sample</u>	Average <sup>a</sup> $R_d$ (m $\mu$ /g)	
	<u>Sorption</u>	<u>Desorption</u>
YM-38	13	120
YM-54	7.8	79
YM-22	2.6	18

<sup>a</sup>Each  $R_d$  value is the average of values obtained from measurements at 3, 6, and 12 weeks with  $<75\mu\text{m}$  and 75- to  $500\mu\text{m}$  fractions.

TABLE IV  
SORPTION RATIOS OF Cs, Sr, Ba, Ce, and Eu UNDER A CONTROLLED  
NITROGEN ATMOSPHERE

<u>Sample</u>	<u>Sorption Time(d)</u>	<u>Desorption Time(d)</u>	$R_d$ (m $\mu$ /g) <sup>a</sup>				
			<u>Cs</u>	<u>Sr</u>	<u>Ba</u>	<u>Ce</u>	<u>Eu</u>
YM-22	48.6		221(2.4)	49.1(2.8)	268(1.5)	920(13.8)	886(4.5)
YM-22		33.1	310(7.5)	94.1(8.1)	464(9.9)	2 420(16.4)	2 840(8.2)
YM-38	51.3		7 760(5.0)	7 733(5.9)	61 200(10.5)	576(7.6)	782(7.1)
YM-38		33.1	14 800(8.1)	13 500(11.5)	73 500(17.7)	2 660(10.3)	2 780(9.1)
YM-54	51.3		236(2.3)	91.5(1.6)	444(1.6)	383(3.0)	649(3.3)
YM-54		33.1	282(7.5)	117(8.0)	555(7.9)	1 380(8.9)	1 830(7.8)

<sup>a</sup>The values in parentheses are the standard deviations for a single measurement of the  $R_d$  values expressed in percent; they were obtained from the errors associated with the activity measurements, and estimated uncertainties were propagated using the rule of change of variables in a moment matrix assuming independence of the variables.

### C. Plutonium and Americium

The initial experiments of the sorptive behavior of plutonium with tuff under the nitrogen atmosphere have been completed;  $^{237}\text{Pu}$  was used as a tracer with three Yucca Mountain tuff samples. The results for 3-, 7-, and 12-week contact times show  $R_d$  values that are comparable to those observed under atmospheric conditions for tuff samples from Jackass Flats. In each case the altered tuff was found to have the highest  $R_d$  value. Little particle-size effect was observed between  $<75\text{-}\mu\text{m}$  and  $75\text{- to }500\text{-}\mu\text{m}$  fractions. Measurements under atmospheric conditions are planned for the Yucca Mountain samples.

## III. MIGRATION RATE STUDIES

### A. Crushed Rock Columns

Elutions of the columns described in the FY-79 tuff report<sup>1</sup> are continuing. Only  $^{85}\text{Sr}$  on tuff from core JA-37 has since been completely eluted. The  $R_d$  value calculated from the column data was 106  $\text{m}\ell/\text{g}$ , compared to a batch value<sup>2</sup> of  $300 \pm 30 \text{ m}\ell/\text{g}$ . An  $R_d$  value approximately two to three times lower with the column method is in agreement with our previous results.<sup>1</sup> In each of those cases the elution curves showed the symmetric peak shapes generally found with ion-exchange columns, and all of the activity was eluted in a narrow volume range. For several of the columns still being eluted a more complex situation is evident. For example,  $^{85}\text{Sr}$  and  $^{137}\text{Cs}$  on material from core JA-18 are showing a slow, "peakless," uniform rate of elution; 7.6%  $^{85}\text{Sr}$  and 7.3%  $^{137}\text{Cs}$  have been eluted in the first 80  $\text{m}\ell$ .

Because the sorption ratios measured for uranium(VI) are small, the retardation factor is expected to be small. With the use of the columns now used in this laboratory, the volume of eluate of interest will be so small as to require drop collection. A method to perform this has been devised using a syringe pump (Sage model 352) and a fraction collector (ISCO model 1200) used in the drop mode. The equipment is contained in a high-humidity box to minimize evaporation of the drops as they form. Aluminum planchets have been made to fit the collection holders. The columns are to be loaded with  $^{131}\text{I}$  and  $^{237}\text{U}$  simultaneously. This will eliminate any uncertainties caused by different conditions during

determination of free column volume and radioisotope collection. The  $^{131}\text{I}$  and  $^{237}\text{U}$  activities on each planchet (1 drop) will be determined using a Ge(Li) detector in conjunction with an automatic sample changer. The 208-keV gamma ray from  $^{237}\text{U}$  and the 364-keV gamma ray from  $^{131}\text{I}$  are readily analyzed. Some difficulty has been encountered due to the tendency of iodide to be lost when dried in carrier-free form. Experiments are in progress putting various "fixers" ( $\text{Na}_2\text{SO}_3$  plus  $\text{AgNO}_3$  and iodide salts) on the planchets before collection of the eluate.

#### B. Infiltration of Rock Cores

The equipment and general technique being used to force solutions of radionuclides into or through solid rock cores and the microautoradiographic (MAR) technique subsequently used to examine those cores are described in Ref. 1. In June 1979,  $^{85}\text{Sr}$  tracer was placed on the top surface of a 5/8-in.-high by 1-in.-diam core of Yucca Mountain tuff sample YM-22. Filtered, rock-equilibrated ground water has been pumped through the core since that time at an average rate of 0.55 ml/day. Residence time of water on the rock column has probably been at least 3 days. A small amount of activity (0.6%) was eluted early (first 2.6 ml) at a decreasing rate. During the next 60 days an additional 0.7% of the strontium was eluted. Since that time the concentration of  $^{85}\text{Sr}$  in the water has increased gradually in an essentially linear fashion; however, the total amount removed in 161 days (89 ml) was only 9.8%. Since the intent of this experiment was to elute the strontium, it will be continued.

By the same technique,  $^{241}\text{Am}$  is being infiltrated into a core of Yucca Mountain tuff sample YM-45 to study porous flow and into a cracked, Climax Stock granite core to study fracture flow. After ground water has been passed through these cores for several weeks, they will be sectioned to examine the sorptive behavior using MAR and counting methods.

#### IV. STATE OF AGGREGATION OF AMERICIUM

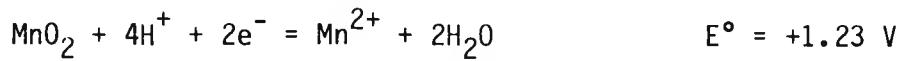
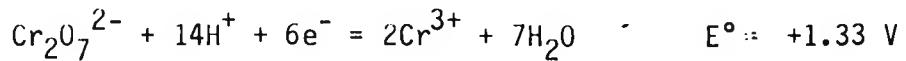
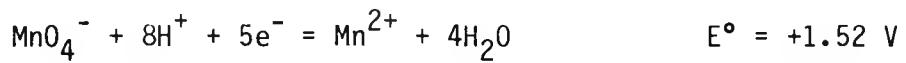
A MAR technique used to investigate retention of americium on filter membranes was described previously.<sup>1</sup> Studies of the "sorption" membranes had shown<sup>1</sup> that not only was americium uniformly sorbed on the filter but also that americium species in a large range of particle sizes were retained. Most "desorption" membranes, however, showed only single alpha

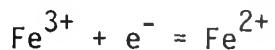
tracks, suggesting that large particulates were not removed from the solids (assuming they had been sorbed) nor were they formed in the aqueous phase during contact. Because it was not known if any of the complex species or polymers that were found to be present in the sorption solutions had been sorbed and were thus "available" to the desorption solutions, the MAR technique was used to look at the crushed rock itself, after completion of the desorption experiments. In five rocks samples examined, all had "clusters" present indicating that large species were indeed sorbed; however, only in one case did the corresponding desorption solution indicate the (minor) presence of clusters.

## V. MATERIALS CHARACTERIZATION

### A. Determination of Iron(II) in Silicate Rocks

The presence of Fe(II) and Fe(III) has important effects on the oxidation potential in geological systems. Total iron can easily be determined. However, a good method for determination of Fe(II) in silicate rocks must involve careful techniques to keep the Fe(II) from oxidizing during preparation, dissolution, and determination. In most procedures, hydrofluoric acid is used; this restricts the container composition. The method that we have selected is based on a procedure<sup>4</sup> in which the silicate rock is dissolved with the simultaneous oxidation of Fe(II) with ICl to form I<sub>2</sub>. This is immediately extracted into carbon tetrachloride, which is present in the dissolving vessel. Iodine monochloride has a lower oxidation potential than either permanganate or dichromate. The use of ICl as oxidant precludes interference by Mn(II) or Cr(III). Pertinent half-cell reactions are





$$E^\circ = 0.77 \text{ V}.$$

The method described above has been modified and tested on several USGS "standard" rocks. The results (see Table V) were usually within 2% of the stated Fe(II) concentration. The  $I_2$  was determined by titration with a standardized  $KIO_3$  solution under conditions where the carbon tetrachloride containing the iodine was kept from contact with the air.

The Fe(II) content will be determined for many of the tuffs already studied: rock samples from studies performed under atmospheric and controlled atmosphere conditions, rock samples and samples pretreated with ground water, and rock samples ground with and without oxygen being present.

#### B. Determination of Iron(II) in Ground Water

Knowledge of the Fe(II)/Fe(III) concentration ratio of solutions that have been pretreated with rocks under various conditions would help in the interpretation of sorption results for multivalent elements to help to substantiate Eh measurements. A method modified from that of Lee and Strumm<sup>5</sup> is being used. The limit of detection is about 0.2  $\mu\text{g}/\text{mL}$ . The small interference of Fe(III) is corrected for. The 4,7-diphenyl-1,1,-10-phenanthroline reagent used is more sensitive and the color developed is more stable than other phenanthrolines. Iron-free reagents needed have been prepared and preliminary measurements are being attempted.

TABLE V  
Fe(II) ANALYSES ON USGS<sup>a</sup> "STANDARD" ROCKS

	% FeO		%
	LASL <sup>b</sup>	USGS <sup>a</sup>	
DTS-1 (Dunite)	7.26	7.23	+0.4
PCC-1 (Peridotite)	5.11	5.24	-2.5
BCR-1 (Basalt)	8.81	8.80	+0.1
GSP-1 (Granodiorite)	2.23	2.31	-3.5
AGV-1 (Andesite)	2.02	2.05	-1.5

<sup>a</sup>USGS = U.S. Geological Society

<sup>b</sup>LASL = Los Alamos Scientific Laboratory

### C. Determination of Redox Potentials in Ground Water

The method used by Grenthe<sup>6</sup> is being tested for the use in measurement of redox potentials. Salt bridges have been constructed for measurement of Eh with a gold electrode.

We will make measurements of the redox potentials of waters under anoxic and atmospheric conditions and after equilibration with tuffs under investigation.

## VI. FIELD TESTS OF RADIONUCLIDES MIGRATION IN GEOLOGIC MEDIA

A major need of the overall National Waste Terminal Storage (NWTS) program to identify and understand the processes that govern radionuclide transport in geologic media is to determine if laboratory studies are being performed under conditions appropriate to natural systems and if models of nuclide transport derived from laboratory results correctly handle element movement under natural conditions.

Indication has been received that the Office of Nuclear Waste Isolation (ONWI) will support a proposal from the Los Alamos Scientific Laboratory (LASL), the Sandia National Laboratories (SNL), and the Argonne National Laboratory (ANL) that addresses these problems. The lead technical organization and principal spokesman for these experiments is LASL Group CNC-11. The program has three principal objectives: (1) to develop the experimental, instrumental, and safety techniques necessary to conduct small-scale radionuclide migration experiments in the field; (2) to use these techniques to define radionuclide migration through rock by performing in situ, at-depth experiments under closely controlled conditions; and (3) to determine whether available material properties and existing or developed models are sufficient to describe real field conditions (that is, to scale from the laboratory to bench size studies to the field).

These tests will be performed in tuff exposed in G-tunnel at the Nevada Test Site (NTS). This site was selected because of the accessibility of formations that allow comparison of laboratory and field measurements and testing of models; the support facilities are already available, mining and drilling costs will be minimized and because radioactive tracers can be used at the NTS with minimal environmental impact. Since G-tunnel is not a potential nuclear waste repository, the experiments will

not compromise the integrity of an actual site. The experiments will concentrate on the migration of selected radionuclides, including actinides, along natural flow paths.

## VII. MINERALOGICAL-PETROLOGICAL STUDIES

Mineralogical and petrological studies were conducted at a reduced level pending availability of new core samples from the Yucca Mountain drilling exploration program. A Siemens automated powder diffractometer is currently being installed and existing x-ray diffraction facilities are being consolidated at a new laboratory location. The new powder diffractometer is operational in the manual mode with installation and alignment of the x-ray generator and diffraction goniostat. The logic controller and computer software are currently being installed. X-ray diffraction patterns for powder samples of core from drill hole UE25a-1 were rerun with the new equipment to recheck previous results and to aid in calibration of the new instrumentation. X-ray powder patterns of differing size fractions of core samples were studied to test for qualitative changes in mineral phases. Only very minor variations were noted. Three high-temperature muffle furnaces were installed in the new laboratory. The furnaces will be used for studies of zeolite stability with temperature.

A contract was let to Professor David Krinsley of Arizona State University to study cathodoluminescence of zeolite minerals in the UE25a-1 drill core. The object of the study is to explore experimental techniques for rapid determination of zeolite chemistry. A contract was let to Professor Paul Knauth, also of Arizona State University, for determination of hydrogen-deuterium ratios in gases evolved from zeolites on heating. The object of the study is to characterize waters derived from different sites or bond-types upon heating of the zeolites.

## VIII. HYDROTHERMAL SOAK TESTS (G-6)

The hydrothermal soak tests are designed to evaluate the effects of extended exposure of selected types of tuff to simulated repository conditions. Tuff samples from UE25a-1 will be subjected to temperatures of 50, 150, and 250°C, confining pressures of 10 and 20 MPa and pore water

TABLE VI  
PARAMETER MATRIX FOR HYDROTHERMAL SOAK TESTS

For each lithologic type of tuff (minimum of two).  
 $P_c$  = confining pressure;  $P_p$  = pore pressure.

Temp	Pressure +	$P_c = 100$ bars	$P_p = 5$ bars	$P_c = 200$ bars, $P_p = 50$ bars
50°C		Basic Parameters <sup>a</sup>		Basic Parameters
		Thermal Parameters <sup>c</sup>		
150°C		Basic Parameters		Basic Parameters
		Permeability (4) <sup>b</sup>		
250°C		Basic Parameters		Basic Parameters <sup>d</sup>
		Thermal Parameters		Permeability (4)

<sup>a</sup>Basic parameters to be measured on all specimens both before and after 6-month soak: tensile strength (Brazil), compressive strength, mineralogy/petrology, density, porosity.

<sup>b</sup>Permeability to be measured on two "before" samples at each T, P indicated. Permeability to be measured after soak at the T, P of the soak--total of four measurements per T, P combination indicated.

<sup>c</sup>Thermal Parameters include thermal conductivity and thermal expansion.

<sup>d</sup>Additional tests will be run at  $P_p = 30$  bars to study effects of water vapor (no permeability measurements).

pressures of 0.5 and 5 MPa for periods up to 6 months (Table VI). Comparative studies of mineralogical and thermomechanical properties will be made before and after exposure. Sample preparation of core from the Topopah Member of the Paintbrush Tuff and the tuffaceous beds of Calico Hills is under way. Ten core samples and one sample of water from Test Well J-13 will be submitted for  $^{18}O$  analysis. All core samples will be characterized petrographically and mineral phases analyzed through use of the electron microprobe and x-ray diffractometer. Data acquisition

systems and controller microprocessor have been procured and software systems for the soak tests are 50% complete. Some difficulty has been encountered in developing a jacketing technique to separate pore water from the confining pressure gas.

Additional work is being conducted to determine the creep response of selected tuff samples from NTS to elevated temperature, confining pressure, and pore water pressure. Correlations between creep response and mineralogic changes will be determined and constitutive relations formulated. A new creep testing apparatus has been designed with a computer-controlled stress actuator. Stress is applied to a sample by a 500 000-lb hydraulic ram pressurized by an air/oil, bladder-type accumulator. Air pressure to the accumulator is adjusted by either adding small volumes of high-pressure air or dumping small volumes. The sign and rate of air-pressure change is controlled by computer actuated solenoid valves based on comparison of current stress on the sample to the desired stress. Stress is determined from the force measured by a load transducer and calculated with corrections for change in cross sectional area of the deforming sample. Currently most components are received or are on order. Construction will begin shortly.

## IX. VOLCANIC HAZARD INVESTIGATIONS

### A. Regional Field Investigations

Geologic field mapping of Plio-Pleistocene volcanic centers in the Reveille and Pancake ranges is continuing (Fig. 1). Basaltic volcanic centers located in the northeastern part of the Reveille Range consist of moderately to deeply dissected cinder cones flanked by lava flows. Conduit plugs and associated dike and sill bodies are exposed within the dissected cone complexes. The dikes vary in thickness from less than 1 m to approximately 10 m and are distributed both radially and concentrically with respect to central conduit plugs. Maximum lateral extent of dikes measured from central conduit plugs is 2 km.

Individual cone centers in the Reveille Range define northeast-trending en echelon zones, suggesting structural control of vent locations by northeast-trending rift zones or faults. Magnetic polarity determinations on basalts in the northeastern Reveille Range have shown that the

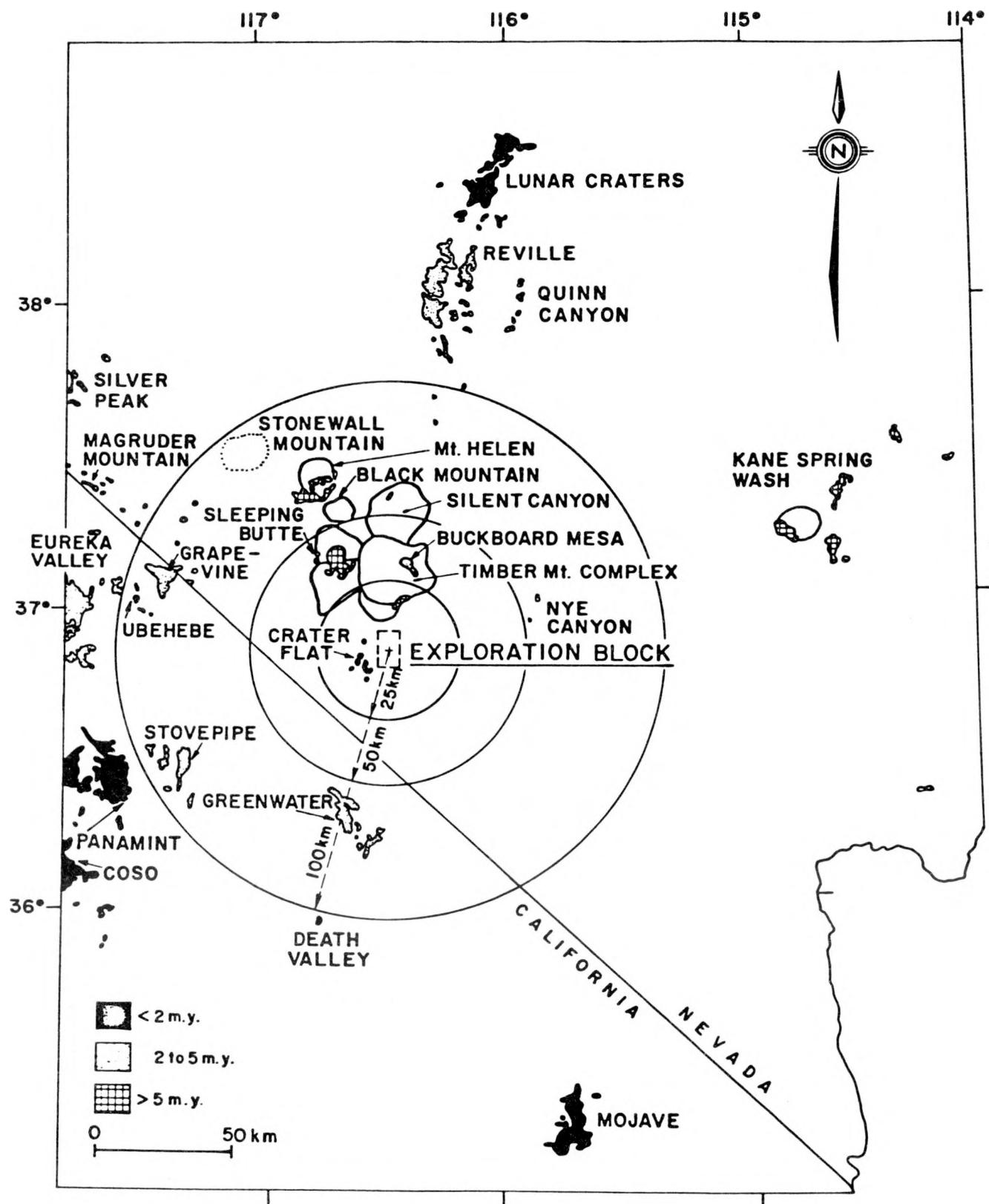


Fig. 1. Distribution of Plio-Pleistocene volcanic centers within the south-central Great Basin. The 25-km, 50-km, and 100-km radius circles were drawn with the center located at the Yucca Mountain exploration block.

basalts are predominantly reversed. Samples will be submitted for age determinations, to correlate K-Ar ages with the magnetic time scale.

Geologic mapping in the Pancake Range was conducted in the central part of the Lunar Crater volcanic field including the volcanic cone clusters of Easy Chair Crater and the Black Rock Summit crater complex. These cone clusters were mapped and the eruptive stratigraphy determined. There appears to be a recognizable trend in nodule and xenocrysts assemblages through time at Easy Chair Crater. Oldest eruptive deposits contain a varied assemblage of lherzolite, clinopyroxenite, and wherlite nodules with megacrysts of olivine, green to black clinopyroxene, and plagioclase. Younger deposits contain apparently increasing amounts of gabbroic xenoliths in addition to the above nodules and xenocrysts suites, with rare amphibole (probable kerasutite).

Eruptive centers of the Lunar Crater volcanic field are strongly aligned along northeast-trending en echelon zones of probable structural weakness. The volcanic field is elongate in a northeast direction along an axial rift zone that has broken across older (Oligocene- and Miocene-age) cauldron complexes that were mapped and described by the U.S. Geological Survey. This axial rift zone is an area of lower topography within the topographically high horst comprising the Pancake Range.

Magnetic polarity determinations of basalts throughout the Lunar Crater volcanic field are consistently normal which suggests that all the basaltic rocks of the field may be younger than 700 000 years. This interpretation will be tested by future age determinations.

#### B. Probability Calculations

Studies of the major parameters controlling calculations of the annual probability of disruption of a repository site by volcanism at Yucca Mountain have been completed. Data used for the calculations are still being obtained but probability bounds for the southern Great Basin can be examined. Rates of volcanic events for the last 2 Myr in the NTS region, based on existing field and geochronology studies, are on the order of  $10^{-8}$  to  $10^{-10}/\text{km}^2/\text{yr}$  which can be expressed as a Quaternary-age cone density of  $10^{-3}$  to  $10^{-4}/\text{km}^2$ . Rates of volcanism for the entire northeast-trending volcanic belt that extends from southern Death Valley, through the Nevada Test Site to the Lunar Crater volcanic field (Fig. 1) are  $10^{-8}$  to  $10^{-9}/\text{km}^2/\text{yr}$ . Maximum rates of volcanism within the volcanic belt were

calculated for the Lunar Crater volcanic field and correspond to  $10^{-7}/\text{km}^2/\text{yr}$  (cone density of about  $0.1/\text{km}^2$ ). For comparison, very approximate rates of volcanic events of  $10^{-9}/\text{km}^2/\text{yr}$  were calculated for the southern Great Basin. The area for this calculation did not include the marginal volcanic provinces of the southern Great Basin but does include the northeast-trending volcanic belt. Excluding the interior volcanic belt yields calculations of about  $10^{-10}$  to  $10^{-11}/\text{km}^2/\text{yr}$ . The area of this calculation, as delineated without the interior volcanic belt, is not generally considered to be an area of active volcanism. Note that the calculated rates of Quaternary volcanic activity for the NTS region overlap with calculated rates of volcanism for this "inactive" area. Comparisons of rates of volcanism cannot be made for marginal volcanic provinces of the Great Basin due to a lack of field and chronology data for volcanism within these provinces.

The disruption probability values for the NTS region are  $3 \times 10^{-8}/\text{yr}$  for a 25-km radius circle (centered at Yucca Mountain),  $1.0 \times 10^{-8}/\text{yr}$  for a 50-km radius circle, and  $4 \times 10^{-9}/\text{yr}$  for a 100-km radius circle (Fig. 1). The minimum or smallest possible probability value approaches but does not reach  $10^{-10}/\text{yr}$  due to the fact that as the radius of the circle of interest is extended beyond 100 km, the area intersects the Lunar Crater volcanic field. This greatly increases the rate calculation, due to the large number of Quaternary cones in the Lunar Crater volcanic field, and the probability values increase to  $10^{-8}/\text{yr}$ . Therefore the probability of disruption of a repository site at Yucca Mountain by future volcanic activity is bracketed by the range  $10^{-8}$  to  $10^{-10}/\text{yr}$ .

### C. Petrography-Geochemistry

The oldest basalts of Crater Flat contain glomeroporphyritic clots that include variable proportions of clinopyroxene and plagioclase. Clinopyroxenes within these clots may have formed during an early phase (at depth?) of crystallization. Clinopyroxenes in the glomeroporphyritic clots have Al/Ti ratios of 6.5:1 that contrast with Al/Ti ratios of about 3:1 in late-stage, groundmass clinopyroxene. These differences in Al/Ti ratios may be caused by a number of factors, all of which strongly suggest differing conditions of crystallization before eruption. This is further supported by calculations based on Na, Al and Si partitioning between plagioclase and rock compositions, assuming that the rock composition

represents the melt composition at the time that the most Ca-rich plagioclase formed. These partitioning functions yield temperature estimates of  $1200^{\circ}\pm75^{\circ}\text{C}$  for a near-liquidus appearance of plagioclase for all of the basalts of Crater Flat, except the glomeroporphritic basalts. The  $1200^{\circ}\text{C}$  temperature estimate is supported by olivine-melt partitioning of Mg and Fe that yield temperature estimates of about  $1125\text{-}1200^{\circ}\text{C}$ . Temperature estimates for plagioclase and olivine should be similar assuming both minerals form at near-liquidus temperatures. However, temperature estimates for plagioclase grains in the glomeroporphritic clots are anomalously high (about  $1350^{\circ}\text{C}$ ). This suggests that either the plagioclase in the clots crystallized at high  $\text{pH}_2\text{O}$ , or that the plagioclase is xenocrystic with respect to the rock in which it now resides. Higher  $\text{pH}_2\text{O}$ , a condition more likely to have occurred uniformly throughout the basalt, is suggested by the uniform distribution of glomeroporphritic clots throughout their host basalts.

Phenocryst-melt or mineral clot-melt element distributions can be used to unravel a multistage basaltic history. These investigations are still in progress for the basalts of Crater Flat. Similar studies will be undertaken for basalts of the Reveille Range that have phenocryst contents that average 14% and are as great as 40%. In comparison, the phenocryst content of the basalts of Crater Flat have a range of only 1-10% and average of 3%.

## X. QUALITY ASSURANCE (CMB-QA)

Implementation of the LASL Quality Assurance Program was continued this quarter. Surveillance of assigned subtasks was performed and documented.

The following quality procedure was drafted, reviewed, approved, and distributed to Nevada Nuclear Waste Storage Investigation (NNWSI) personnel: TWS-CMBQA-QP-04, R0, Handling, Storage and Shipping Procedure.

Drafts of the following work plans and procedures were written, reviewed and are in the process of being finalized:

TWS-G6-WP-07, RO	Hydrothermal Soaking Experiments
TWS-G6-DP-09, RO	Density and Porosity Determinations
TWS-G6-DP-10, RO	Compressive Strength Tests
TWS-G6-DP-11, RO	Brazil Indirect Tensile Strength Test

The Personnel Certification form was finalized and certifications for LASL personnel working on the NNWSI program are now complete and on file.

LASL quality assurance personnel met with USGS personnel on December 12 to discuss plans for the continued development and implementation of the USGS Quality Assurance Program.

#### REFERENCES

1. E. N. Vine, R. D. Aguilar, B. P. Bayhurst, W. R. Daniels, S. J. DeVilliers, B. R. Erdal, F. O. Lawrence, S. Maestas, P. Q. Oliver, J. L. Thompson, and K. Wolfsberg, "Sorption-Desorption Studies on Tuff II. A Continuation of Studies with Samples from Jackass Flats, Nevada and Initial Studies with Samples from Yucca Mountain, Nevada," Los Alamos Scientific Laboratory report LA-8110-MS (1980).
2. K. Wolfsberg B. P. Bayhurst, B. M. Crowe, W. R. Daniels, B. R. Erdal, F. O. Lawrence, A. E. Norris, J. R. Smyth, "Sorption-Desorption Studies on Tuff I. Initial Studies with Samples from the J-13 Drill Site, Jackass Flats, Nevada," Los Alamos Scientific Laboratory report LA-7480-MS (1979).
3. B. M. Crowe (compiler), "Research and Development Related to the Nevada Nuclear Waste Storage Investigations, July 1-September 30, 1979," Los Alamos Scientific Laboratory report LA-8152-PR (1979).
4. S. Banerjee, Anal. Chem. 46, 762 (1974).
5. G. F. Lee and W. Stumm, Jour. AWWA 52, 1567 (1960).
6. I. Grenthe, "Determination of Redox Potential in Ground Water from Stripa and Finnsjo," in "Copper as Canister Material for Unreprocessed Nuclear Waste - Evaluation with Respect to Corrosion," KBS Teknisk Rapport 90 (1978), Swedish Corrosion Research Institutue, Appendix B5.
7. M. J. Drake, "Plagioclase-melt Equilibria," Geochm. Cosmochim. Acta. 40, p. 457-466.

## PUBLICATIONS

- B. M. Crowe and W. J. Carr, "Preliminary assessment of the risk of volcanism at a proposed nuclear waste repository in the southern Great Basin: U.S. Geol. Surv. Open File Report (in press).
- M. L. Sykes, J. R. Smyth, and G. H. Heiken, "Zeolite mineralogy and chemistry in some Tertiary Tuffs of Southern Nevada, EOS (in press).

## PRESENTATIONS

- B. R. Erdal, B. P. Bayhurst, W. R. Daniels, S. J. DeVilliers, G. M. Heiken, F. O. Lawrence, M. L. Sykes, J. R. Smyth, J. L. Thompson, E. N. Vine, and K. Wolfsberg, "Parameters Affecting Radionuclide Migration in Geologic Media," Symposium on the Scientific Basis for Nuclear Waste Management, Materials Research Society, Boston, MA, November 26-29, 1979.
- J. R. Smyth, "Zeolites and radioactive waste isolation," talk presented at the Geology Department, University of California, Santa Barbara.
- M. L. Sykes, J. R. Smyth, and G. H. Heiken, "Zeolite mineralogy and chemistry in some Tertiary Tuffs," American Geophysical Union Fall Meeting, San Francisco, CA.

Printed in the United States of America. Available from

National Technical Information Service

US Department of Commerce

5285 Port Royal Road

Springfield, VA 22161

Microfiche \$3.00

001-025	4.00	126-150	7.25	251-275	10.75	376-400	13.00	501-525	15.25
026-050	4.50	151-175	8.00	276-300	11.00	401-425	13.25	526-550	15.50
051-075	5.25	176-200	9.00	301-325	11.75	426-450	14.00	551-575	16.25
076-100	6.00	201-225	9.25	326-350	12.00	451-475	14.50	576-600	16.50
101-125	6.50	226-250	9.50	351-375	12.50	476-500	15.00	601-up	

Note: Add \$2.50 for each additional 100-page increment from 601 pages up.