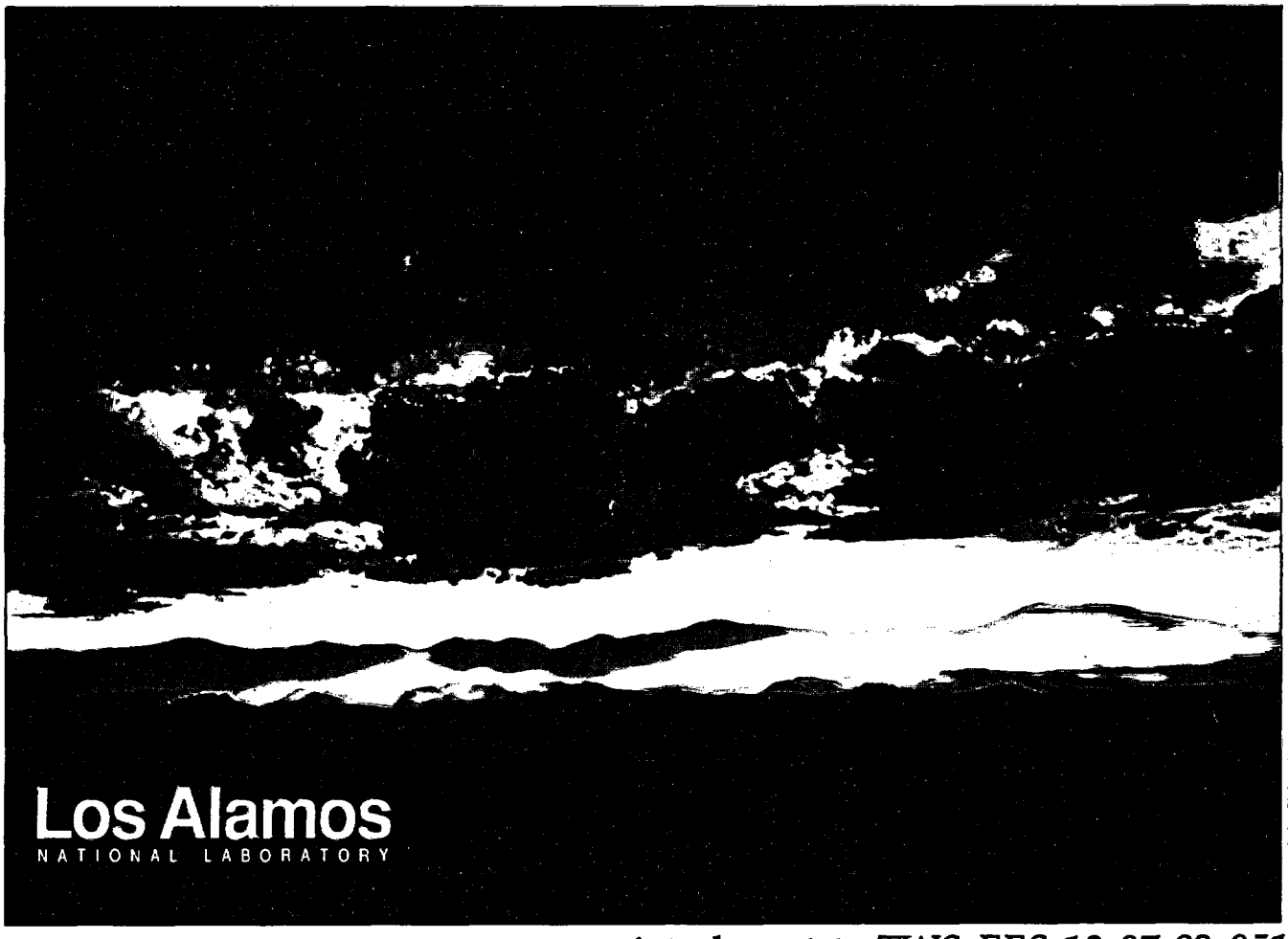
Yucca Mountain Site Characterization Project

Monthly Activity Report

June 1992



Photograph: by Chris J. Lindberg

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LOS ALAMOS NATIONAL LABORATORY

YUCCA MOUNTAIN SITE CHARACTERIZATION PROJECT

Monthly Activity Report
June 1992

WBS 1.2.1 Systems

Objective

The objective of this task is to integrate systems with the Geologic Repository Program, to describe the Yucca Mountain Site Characterization Project Mined Geologic Disposal System, and to evaluate the performance of the natural, engineered barrier, and total systems for meeting regulatory standards.

Technical Data (WBS 1.2.1.3.5)

Activities and Accomplishments

Finalized Quality Administrative Procedure 8.3, "Transfer of Data."

Staff entered data into the Automated Technical Data Tracking System for Activities 8.3.1.8.1.3.2, "Preliminary Geologic Map of Sleeping Butte Volcanic Centers"; 8.3.1.3.6.1.1, "Radionuclide Migration Laboratory Studies" (6); and 8.3.1.3.2.1.3, "Fracture-lining Manganese-Oxide Minerals in Silicic Tuff."

Planned Activities

Submit data from Activities 8.3.1.8.1.3.2, 8.3.1.3.6.1.1 and 8.3.1.3.2.1.3 to the Technical Data Base.

Work on Technical Data Base parameter screens.

Caisson Experiment (WBS 1.2.1.4.6)

Activities and Accomplishments

Filling of the caisson is awaiting the NEPA categorical exclusion from DOE Albuquerque Operations Office.

A design to simulate the lower-boundary condition has been supplied by R. Glass of Sandia National Laboratory. This design uses approximately 300 suction candles mounted on a plastic plate, and tubing from the candles will be routed through PVC pipe to the central access caisson. Technicians at Los Alamos will fabricate the unit with guidance from Glass.

Before preliminary modeling activities can begin, the hydrologic properties of the sand and sand/limonite mixture must be determined. This phase of the characterization will be contracted.

Continued on next page

Planned Activities

Prepare the caisson for filling.

Construct lower-boundary condition apparatus as soon as material becomes available. Revise SOP as dictated by review.

Write manuscript for special issue of *Radioactive Waste Management.*

Publications

E. P. Springer and M. D. Siegel
An Integrated Intermediate-Scale Caisson Experiment to Validate Models of Fluid Flow and Contaminant Transport in the Unsaturated Zone
Journal article, *Radioactive Waste Management and the Nuclear Fuel Cycle - Special issue on the Yucca Mountain Project*
In preparation.

Performance Assessment Calculational Support (WBS 1.2.1.4.7)

Activities and Accomplishments

Work continued on a three-dimensional finite-element mesh model for study of radionuclide transport using the dual porosity/dual permeability capability (transporting nuclides through the matrix and fractures separately) available in FEHMN. The results will be compared to results from analogous calculations using the composite-porosity model.

Planned Activities

Continue to generate three-dimensional finite-element mesh model.

WBS 1.2.3.1 Site Management and Integration

Site Management (WBS 1.2.3.1.1)

Objective	The objective of this task is to manage and integrate site characterization activities.
Activities and Accomplishments	Management staff continued intensive work on Mission 2001, hosted internal QA audits, participated in ITE, and had progress meetings with key technical staff and supervisors.

Test Management and Planning (WBS 1.2.3.1.2)

Objective	The goal of this investigation is to provide coordination for Los Alamos surface-based test planning package development.
Activities and Accomplishments	The development of design and test-related information (D&TRI) submissions were begun for the UZ-14, C-Well, and the neutron drilling program test planning and job packaging. Staff began developing information for tests to be performed in the north-portal area. Staff will also begin to prepare test planning packages for launch chamber tests. Specifications for a photographic indexing system were written and transmitted to the Project office. Sample collection instructions were written for future UZ and neutron drilling operations by J. Fabryka-Martin, principal investigator of Task 1.2.3.3.1.2.2, and submitted to the Project office. Several sample requests were evaluated and approved by the SOC.
Planned Activities	Continued support of the los Alamos surface-based site characterization activities in response to the Project program directives.
Problem Areas	None
Milestones	None

WBS 1.2.3.2.1.1.1 Mineralogy, Petrology, and Rock Chemistry of Transport Pathways

Objective

The purpose of this activity is to define the important mineralogic and geochemical variables along fracture and rock-matrix transport pathways at Yucca Mountain, in support of performance assessment and to evaluate the impact of repository construction on natural waste-transport barriers.

Activities and Accomplishments

We are developing operating procedures for using the INEL microdiffractometer. A program is being written to convert the INEL data to Siemens DIFFRAC 5000 format, which will permit us to use existing software to analyze the microdiffractometer data.

A first draft of "Calcite deposits in fractures of drill cores USW G-2 and USW GU-3/G-3 at Yucca Mountain, Nevada" (milestone 3120) has been completed. This paper summarizes chemical and petrographic data for samples from which twelve calcite separates were prepared for instrumental neutron-activation analysis (INAA).

Author modifications have been compiled for a report, "Geologic evaluation of six nonwelded tuff sites for a surface-based test facility for the Yucca Mountain Project" (milestone 3137); the authors are D. Broxton, S. Chipera, and F. Byers. This paper combines x-ray fluorescence (XRF), atomic absorption (AA), phosphomolybdate colorimetry (PMC), INAA, x-ray diffraction (XRD), and thin-section modal data on surface samples to compare six potential localities for a surface-based test facility. The authors recommend nonwelded vitric tuffs at Busted Butte and/or zeolitic tuffs at Prow Pass as the most suitable locations for a surface-based test facility to investigate tuff properties in rock analogous to the tuffaceous beds of Calico Hills beneath Yucca Mountain.

B. Carlos attended the joint Geochemistry/Hydrology Integration Meeting on "Fast Pathways: Definition and Detection" in Denver, CO on 9-11 June. She presented a summary of her work on fracture mineralogy relevant to the transport-pathway definition and participated in the working group for saturated-zone transport.

Planned Activities

Work planned within the next few months includes the following activities: (1) continue analysis of fracture fillings in the Paintbrush Tuff to determine mineral distribution and factors controlling that distribution and (2) continue analysis of calcites to understand transport and precipitation mechanisms.

Problem Areas

Milestone 3120 has been delayed because of (1) redirection of a larger part of the author's effort into calcite-silica studies under 1.2.3.2.1.1.2 and (2) expansion of the scope of the milestone to include more data on calcites below the water table.

Milestone Progress

3120
30 June 1992
Calcite in the Upper Paintbrush Tuff
95% complete.

3130
17 August 1992
Fracture Mineralogy of the Paintbrush Tuff

Continued on next page

3137
30 September 1992
Mineralogy of Calico Hills for Adit Development
99% complete.

Publications

D. E. Broxton
Chemical Changes Associated with Zeolitization on the Tuffaceous Beds of Calico Hills at Yucca Mountain, Nevada
Conference paper, *Proceedings of the 7th Water-Rock Interactions Symposium*, July 1992
Approved by YMPO.

B. Carlos, D. Bish, S. Chipera, and S. Craven
Fracture-Lining Manganese Oxide Minerals in a Silicic Tuff
Journal article, *Chemical Geology*
Submitted to YMPO.

G. D. Guthrie, D. L. Bish, and B. T. Mossman
Quantitative Analysis of Zeolite-Bearing Dusts Using the Rietveld Method
Journal article, *Science*
Submitted.

D. Vaniman, D. Bish, D. Broxton, B. Carlos, S. Chipera, and S. Levy
Mineralogy as a Factor in Radioactive Waste Transport Through Pyroclastic Rocks at Yucca Mountain, Nevada
Journal article, *Journal of Geophysical Research*
Submitted to TPO.

WBS 1.2.3.2.1.1.2 Mineralogic and Geochemical Alteration

Objective

The objective of this task is to characterize past and present natural alteration processes that have affected the potential geologic repository and to predict future effects of natural and repository-induced alteration.

Activities and Accomplishments

A paper on pedogenesis of siliceous calcretes at Yucca Mountain by D. Vaniman, S. Chipera, and D. Bish has been revised. The paper is intended for publication in *Science*.

Powders of bedrock, calcite-silica deposits, and other alteration products from Trenches 14 and 14A were prepared for instrumental neutron-activation analysis (INAA).

G. WoldeGabriel examined altered samples using scanning electron microscopy (SEM) to characterize their secondary mineral associations. He is collected textural evidence for the relative timing of mineral dissolution and precipitation.

S. Levy attended the joint Geochemistry/Hydrology Integration meeting in Denver. She presented evidence from alteration history studies pertinent to the identification of "fast" or "preferred" fluid pathways. She presented information on studies of transient hydrothermal systems in the cooling Topopah Spring and Tiva Canyon Members of the Paintbrush Tuff as natural analogs for water infiltrating a hot repository.

Planned Activities

Analysis of samples from Trench 14 and other surface samples will continue. D. Vaniman will research the pedogenic sources of chemical species involved in unsaturated-zone transport at Yucca Mountain. S. Levy will summarize the evidence pertinent to the issue of surface discharge of water from depth.

Mineralogy-petrology researchers plan to meet with USGS representatives in July to discuss cooperative research.

Problem Areas

None

Milestone Progress

3138
30 October 1992
Chemical Transport in Zeolitic Alteration
60% complete.

3141
31 March 1992 (delayed due to participation in the issue resolution process)
Laminated Zone in Trench 14
62% complete. (Technical review complete.)

3142
31 July 1992
K/Ar Dating of Clays and Zeolites
Research continuing; new draft completed.

3143
30 April 1992
Experimental Dehydration of Volcanic Glasses
Submitted to TPO.

Continued on next page

3150
15 April 1993
Final Report on Bedrock
24% complete.

Publications

S. Levy and C. Naeser
Bedrock Breccias Along Fault Zones near Yucca Mountain, Nevada
Chapter in USGS Bulletin on Yucca Mountain studies
In USGS editorial review.

S. Reneau
Maganese Accumulation in Rock Varnish in a Desert Piedmont, Mojave Desert, California, and Application to Evaluating Varnish Development
Journal article, *Quatenary Research*
Submitted to TPO.

D. Vaniman, D. Bish, and S. Chipera
Dehydration and Rehydration of a Tuff Vitrophyre
Journal article, *Journal of Geophysical Research*
Submitted to TPO.

D. Vaniman, *et. al*
Precipitation of Calcite, Dolomite, Sepiolite, and Silica from Evaporated Carbonate and Tuffaceous Waters of Southern Nevada
Conference paper, *Proceedings of the 7th Water-Rock Interactions Symposium*, July 1992
Approved by YMPO.

D. Vaniman, S. Chipera, and D. Bish
Pedogenesis of Siliceous Calcretes at Yucca Mountain, Nevada
Journal article, *Science*
Submitted to TPO

G. WoldeGabriel, *et. al.*
Preliminary Assessment of Clinoptilolite K/AR Results from Yucca Mountain, Nevada: a Potential High-Level Radioactive Waste Repository Site
Conference paper, *Proceedings of the 7th Water-Rock Interactions Symposium*, July 1992
Approved by YMPO.

WBS 1.2.3.2.1.2 Stability of Minerals and Gases

Objective

The objective of this activity is to produce a model for past and future mineral alteration in Yucca Mountain. The model is intended to explain the natural mineral evolution resulting from the transformation of metastable mineral assemblages to more stable assemblages and the effects of a repository emplacement.

Activities and Accomplishments

This activity has been deferred.

WBS 1.2.3.2.5 Postclosure Tectonics

Objective

The objective of these volcanism studies is to determine the hazards of future volcanic activities with respect to siting a high-level radioactive waste repository at Yucca Mountain.

Activities and Accomplishments

Twelve samples were submitted to Lehigh University for $^{40}Ar/^{39}Ar$ age determinations, and thirty-nine samples were submitted to Washington University for instrumental neutron-activation analyses.

Three trenches were completed in the scoria-fall and pyroclastic surge deposits about 1 km northwest of the main cone at the Lathrop Wells Volcanic Center. A distinct group of thin-bedded, pyroclastic surge deposits in two of the trenches was exposed and described. We infer that these deposits correlate with the scoria-fall and pyroclastic surge deposits south of the main cone and beneath the tephra deposits with interbedded soils. There are now 32 trenches at the Lathrop Wells Center.

Three trenches were completed at the "A" Cone of the Cima Volcanic Field. The soil units in the soils were described, and the exposed tephra units were correlated with field and geomorphic units of the center.

Mineral separations were completed for samples from the Ql_6 lava flow at the Lathrop Wells Center and the Western lava flow at the Little Black Peak Center.

We evaluated an alternative method to degas olivine grains for ^3He cosmogenic age determinations. This method allows for both lower blank corrections and higher sample throughput. A new furnace design will be required to use this method.

Work continued on the Issue Resolution Report for volcanism, which will be completed in July.

Work continued on revising Los Alamos standard operating procedures (SOPs) for trenching operations using a truck-mounted backhoe. On-site training was completed, and two staff members became qualified to conduct trenching and/or train others in preparing trenches less than five feet deep.

The restriction on generation of mixed waste for uranium-thorium separation work has been lifted. Four sets of samples are now being dissolved for mass spectrometry studies.

Software certification continues for ^3He mass spectrometry studies. To support submission of volcanism data to the RIB and SEPDB, we are processing field notebooks used before the QA program was instituted.

Planned Activities

A video conference on volcanism with the NRC is planned for August.

A special volume on volcanic chronology and risk assessment with special emphasis on studies of the Lathrop Wells Volcanic Center is planned for a 1993 issue of the *Journal of Volcanology and Geothermal Research.*

Problem Areas

None

Continued on next page

Milestone Progress

3174
8 January 1992
Effects of Magmatic Disruption on the Repository (Study Plan 8.3.1.8.1.2, R0)
Submitted to TPO.

3034
30 September 1992
Report on Magma System Dynamics

3109
30 September 1992
Report of Subsurface Effects

3111
30 September 1992
Preliminary Geologic Mapping of Volcanic Centers

3164
30 September 1992
Progress Report on Thermoluminescence

R482
31 August 1992
Issue Resolution Report

Publications

B. M. Crowe, *et. al.*
Issue Resolution Report
In preparation.

B. M. Crowe, G. Valentine, R. Morley and F.V. Perry,
1992, *Recent Progress in Volcanism Studies: Site Characterization Activities for the Yucca Mountain Project,*
Proceedings paper, *Waste Management92, Proceedings of the Symposium on Waste Management,*
Published.

S. G. Wells, *et al.*
Multiple Eruptive Events at Small Volume Basaltic Centers: Evidence From the Cima and Crater Flat Volcanic Fields
Journal article
In preparation.

WBS 1.2.3.3.1.2.2 Water-Movement Tracer Tests

Objective

The objective of the water-movement tracer tests is to obtain measurements of chlorine isotope distributions to help quantify the percolation of precipitation in the unsaturated zone.

Activities and Accomplishments

The subcontractor, Hydro Geo Chem, completed analyses of chloride and bromide concentrations in selected cutting samples from USW UZ-N54, USW UZ-N55, and USW UZ-N37 and from Midway Valley soil samples to help estimate the quantity of sample required to be leached to ensure sufficient chloride for isotopic analysis. Chloride concentrations in the cutting samples ranged from 3 to 48 ppm, and the approximate average for 25 samples was 5 ppm; only 3 samples had concentrations exceeding 9 ppm. Chloride/bromide ratios were highly variable for these samples; the data are being closely examined to determine the extent to which the variability is attributable to bromide concentrations that are close to the detection limit. The Midway Valley soil samples, which were collected from trenches at depths of 1.5 to 3 m, generally had much higher chloride concentrations, averaging about 100 ppm but ranging up to 700 ppm. All data will be checked carefully.

Analyses of $^{36}Cl/Cl$ were received for water samples from UE-25 UZ n#2, UE-29 UZ-N91, and UE-25 p#1 and for 11 cuttings samples from USW UZ-N54 and N55. In USW UZ-N54, bomb-pulse ^{36}Cl was detected in the alluvium down to a depth of 15-17 feet; below that depth, it was present in the alluvium only at background levels. This distribution is consistent with the moisture profile obtained by A. Flint (USGS Study Plan 8.3.1.2.2.3) for this drill hole, indicating the effectiveness of the alluvium and associated vegetation in reducing the net infiltration rate. Analyses of $^{36}Cl/Cl$ were also obtained for the bedded tuff unit underlying the Tiva Canyon welded unit in USW UZ-N55. Unexpectedly high values throughout this unit may indicate the presence of a "fast path" through the welded unit; however, several of the samples have values that are considerably higher than can be explained by global fallout of ^{36}Cl. Considerable analysis is required to verify whether contamination can be ruled out as the source for these high results. All values are preliminary and require detailed checking to determine their acceptability; no data have as yet been submitted to the Technical Data Base. These data may not be cited until approved by Los Alamos. The data were presented by J. Fabryka-Martin at the Joint Geochemistry/Hydrology Integration meeting 9-11 June in Denver.

The final version of LANL-INC-DP-90, R1, "Measurement of Moisture Content of Soil Samples," was distributed. Technical and QA reviews of LANL-INC-DP-94, R0, "Using Ion Chromatography to Determine Chloride and Bromide Concentrations," were completed and forms are being revised. LANL-INC-DP-87, R2, "Identification, Storage and Handling of Samples at Hydro Geo Chem," was prepared and submitted for technical and QA reviews.

Collection of cutting samples for 36Cl analysis from UZ-16 continued this month.

Planned Activities

Complete additional DPs; process soil samples for Cl/Br and chlorine-36/Cl ratios; process cuttings samples from neutron-access boreholes; collect additional soil samples from Yucca Mountain area as opportunities arise.

Problem Areas

None

Continued on next page

Milestone Progress

3191
Procedure for Chlorine-36 Analysis of Unsaturated Zone Samples
30 September 1992
85% complete

WBS 1.2.3.3.1.2.5 Diffusion Tests in the ESF

Objective	The objective of this task is to determine *in situ* the extent to which the nonsorbing tracers diffuse into the water-filled pores of the Topopah Spring welded unit.
Activities and Accomplishments	No significant activity in this study.
Milestone Progress	No Level II milestones are planned this fiscal year.

WBS 1.2.3.3.1.3.1 Site Saturated Zone Groundwater Flow System (Reactive Tracer Testing)

Objective

Experiments will be conducted at the C-Well complex (holes UE-25 c#1, UE-25 c#2, and UE-25 c#3) and other wells in the vicinity of Yucca Mountain using reactive tracers to characterize retardation and transport properties at a larger scale than currently used in laboratory experiments.

Activities and Accomplishments

Software Certification. We are delaying this effort until new SQA staff are in place. Z. Dash and B. Robinson will serve as temporary Software Configuration Manager and CCB Chair, respectively.

Lithium Bromide Studies. Batch-sorption studies continued; a report on their status will be included in the July report.

Modeling. The dual porosity option of the FEHMN application is being tested so that we may conduct non-quality-affecting design calculations to determine the length of time required to take to establish a steady-state flow field between two of the C-Wells in a recirculating tracer experiment.

Planned Activities

Continue batch-sorption experiments with lithium bromide.

Continue developing techniques for measuring the concentration of polystyrene microspheres in solution.

Complete paper on a validation strategy for the matrix diffusion conceptual model.

Continue modeling studies using FEHMN to support design of field tests.

Problem Areas

Because of the lack of staff assigned to the SQA activity, we are unable to progress complete the certification effort for SORBEQ, FEHMN, and GZSOLVE.

Publications

W. L. Polzer, E. H. Essington
The Use of Selectivity Coefficients to Estimate Modified Langmuir Isotherm Parameters as a Function of Experimental Conditions
Journal article, *Radioactive Waste Management and the Nuclear Fuel Cycle - Special issue on the Yucca Mountain Project*
In preparation.

W. L. Polzer, M. G. Rao, H. R. Fuentes, and R. J. Beckman
Thermodynamically Derived Relationships Between the Modified Langmuir Isotherm and Experimental Parameters
Journal article, *Environmental Science and Technology*
Undergoing revision.

Continued on next page

B. A. Robinson
FRACNET—Fracture Network Model for Water Flow and Solute Transport
LA-series report
In preparation.

B. A. Robinson
SORBEQ—A One-Dimensional Model for Simulating Column Transport Experiments
LA-series report
In preparation.

B. A. Robinson
A Strategy for Validating a Conceptual Model for Radionuclide Migration in the Saturated Zone Beneath Yucca Mountain
Journal article, *Radioactive Waste Management and the Nuclear Fuel Cycle - Special issue on the Yucca Mountain Project*
In preparation.

Milestone Progress

3188
30 September 1992
Documentation for SORBEQ

3194
30 September 1992
Batch Sorption Experiments with Lithium

T112
22 June 1992
Final Documentation for FEHMN
Delayed due to change of assignment of principal investigator.

3196
27 July 1992
FRACNET Documentation

WBS 1.2.3.4.1.1 Groundwater Chemistry Model

Objective

The goal of this investigation is to provide conceptual and mathematical models of the groundwater chemistry at Yucca Mountain. These models will explain the present groundwater composition in relation to interactions of minerals and groundwater and will be used to predict groundwater compositions as a result of anticipated and unanticipated environments.

Activities and Accomplishments

Study Plan. Review comments on the Groundwater Chemistry Model Study Plan, R0, are being addressed by M. Ebinger.

A matrix for investigating most-active groundwaters is being developed. We plan to categorize groundwaters by compositional variables that affect radionuclide solubility and/or soption properties. Variables of interest are the following: measured Eh potential, pH, and bicarbonate content. EQ3/6 will be used to conduct simulations using the different kinds of groundwater compositions.

Gas chromatography of dissolved gases in groundwaters is one technique for determining Eh of waters from Yucca Mountain. At this time, however, we are considering various other methods for measuring CO_2, CH_4, and H_2 in groundwater. Additional groundwater components may be considered at a later date.

QA Activities. No additional progress to report on the IMOU between LLNL and Los Alamos. IMOU is in review in Las Vegas.

Planned Activities

Continue resolution of comments on the Study Plan.

USGS collaboration will continue. Delays in material and equipment for the USGS down-hole sampler could slow progress on testing of conceptual models of groundwater chemistry at Yucca Mountain.

Continue support of QA efforts. Continue tracking IMOU mentioned above.

Problem Areas

None

Milestones Progress

3006
31 August 1992
Eh and pH Buffering Capacity

3415
30 September 1992
Letter Report: *Most-Active Groundwater Chemistry*

Publications

M. Ebinger
Water-Rock Interactions and the pH Stability of Ground waters from Yucca Mountain, Nevada
Conference paper, *Proceedings of the 7th Water-Rock Interactions Symposium*, July 1992
Approved by YMPO.

Continued on next page

D. Vaniman, D. Bish, M. Ebinger, S. Chipera
Precipitation of Calcite, Dolomite, Sepiolite, and Silica from Evaporated Carbonate and Tuffaceous Waters of Southern Nevada
Conference paper, *Proceedings of the 7th Water-Rock Interactions Symposium*, July 1992
Approved by YMPO.

WBS 1.2.3.4.1.2.1/3 Batch-Sorption Studies and Sorption Models

Objective

The objective of this task is to provide sorption coefficients for elements of interest to predict radionuclide movements from the repository to the accessible environment.

Activities and Accomplishments

New staff members K. Kung and J. Schense were oriented to their positions in the YMP Sorption Task.

Staff reviewed the June 1991 - January 1992 BET surface area measurements and decided to repeat them.

Planned Activities

In July, absolute accuracy of the BET apparatus will be determined, a DP for sorption measurements will be prepared, and possible systematic errors due to BET calibration method will be assessed.

S. Kung will prepare his new laboratory space for sorption measurements and write the appropriate Standard Operating Procedures (SOPs).

Problem Areas

None

Milestone Progress

3009
30 September 1992
Variation of Water-Rock Ratio Sorption Coefficients on Zeolitic Tuff

3212
30 September 1992
Progress Report on Single Mineral Experiments

Publications

A. Meijer
A Strategy for the Derivation and Use of Sorption Coefficients in Performance Assessment Calculations for the Yucca Mountain Site
Conference proceedings, *Proceedings of the DOE/Yucca Mountain Site Characterization Project Radionuclide Adsorption Workshop at Los Alamos National Laboratory September 11-12, 1990.* (LA-12323-C, 1992)
In press.

WBS 1.2.3.4.1.2.2 Biological Sorption and Transport

Objective

The purpose of this research is to determine whether microbial activity can influence the movement of plutonium in tuff. Because fluids are used extensively in the exploration of locations for a nuclear repository, those micro-organisms capable of utilizing drilling fluids as growth substrates are of special interest.

Activities and Accomplishments

The dissolution of hematite by the Pseudomonas sp. is being determined. These experiments are being performed to aid in determining the effect(s) of microorganisms on actinide oxides and to better understand the activities of microorganisms in a subsurface environment.

Unsaturated flow columns are now being constructed to study microbial activities in unsaturated environments.

Work has continued on writing milestones 3080, "Report on Chelation," and 3092, "Report on Colloidal Agglomeration."

Planned Activities

Continue plutonium K_d experiments.

Continue colloidal agglomeration experiments.

Problem Areas

None

Milestone Progress

3080
30 September 1992
Report on Chelation
In preparation.

3092
30 September 1992
Report on Colloidal Agglomeration
In preparation.

3176
30 September 1992
Procedure for Determination of Formation Constants
In progress.

3177
30 September 1992
Procedure for Determination of Effects on Colloidal Agglomeration
In preparation.

Publications

L. R. Hersman, D. E. Hobart, and T. W. Newton
Preliminary Evidence of Siderophore/Plutonium Complexation
Journal article, *Journal of Applied and Environmental Microbiology*
Resubmitted.

WBS 1.2.3.4.1.3 Radionuclide Retardation by Precipitation Processes

Objective

The objective of the solubility determination task is to determine the solubilities and speciation of important waste elements under conditions characteristic of the repository and along flow paths from the repository into the accessible environment.

Activities and Accomplishments

We continued to revise the Study Plan and expect to complete it by 15 August.

Speciation Studies. Plutonium carbonate NMR results (milestone 3031) are being written up and will be submitted to the *Journal of the American Chemical Society*. D. Clark has modified "Molecular Models for Actinide Speciation" in response to TPO comments. "Plutonium(IV) Carbonate Speciation Changes" by Tait *et. al.* was submitted for technical review. This paper describes our preliminary PAS results on the Pu(IV) carbonate system and will be submitted to *Inorganic Chemistry*. An abstract, "Speciation of Plutonium(IV) in Carbonate Media," by Morris, *et. al.* was prepared for a symposium entitled "The Scientific Basis for Nuclear Waste Management XVI" to be held at the fall meeting of the Materials Research Society.

To verify earlier data analysis, we performed a complete 13-C NMR experimental relaxation measurement. Am(III) 17-O NMR experiments have begun, and a complete series of Am(III) and Pu(III) NMR experiments was performed.

We have resumed photoacoustic spectroscopy (PAS) data collection on the Pu(IV) carbonate system as the question about possible generation of mixed waste has been resolved. We have begun the certification process for commercial NMR software. Preparation of Np(V) solutions for sorption studies has been postponed until July.

Solubility Studies. The neptunium(Np) undersaturation experiments at pH 6.7 and 8.5 have been started, and the experiment at pH 6 has reached the same soluble concentration that we observed in the oversaturation experiments. The soluble Np concentration at day 85 was within 5% of the final concentration measured in the oversaturation experiment. At pH 7, steady-state was achieved.

The irregular behavior noted in the Np oversaturation experiment at pH 7 showed signs of repeating itself in the undersaturation experiment; however, this irregularity may be caused by the pH variation. The pH values and Np concentrations that correspond to the three most recent samplings of the pH 7 experiment are shown below:

Assay #	Day	pH	Np concentration
2-16AIU6	15	7.05	3.4×10^{-5} M
2-17AIU6	42	6.93	6.4×10^{-5} M
2-18AIU6	72	7.23	4.2×10^{-5} M

Np shows a great change in solubility as a function of pH: any deviation below pH 7 will tend to make the Np more soluble, and any deviation above pH 7 will make the Np less soluble. Maintaining a neutral pH is very difficult, and this effect can be seen in the assays of the pH 7 experiment on days 42 and 72. The higher pH produced a lower concentration, which is effectively what we observed across the three Np experiments. Because pH deviation is already being controlled as closely as possible, we plan to monitor the correlation between concentration and pH in the pH 7 experiment more closely in the future.

Continued on next page

For the experiment at pH 7 and 8.5, we found that computer-controlled monitoring compensated for frequent pH fluctuations by adding volumes of acid or base to return the pH to the desired level; by the time the oversaturation experiments had reached 100 days of reaction time, the experiments at pH 7 and 8.5 had 30 and 75% respectively of their initial starting volume added. To prevent this from happening in the future, we have stopped the automated pH adjustment.

The pH 6 experiment was much more stable. At days 85, 72, and 100, we added no more than 2 mL of acid and base by hand to the cells, only about 3% of the initial volume. By comparing the two approaches (oversaturation and undersaturation) at pH 8.5 we see that the dilution on UE-25 p#1 by the addition of acid and base shows no significant effect on the solubility of Np. After 100 days of reaction time, the pH 8.5 undersaturation experiment virtually reproduced the oversaturation experiment at pH 8.5.

Results of the x-ray powder diffraction studies on the solids produced in the three Np oversaturation experiments will included with next month's letter report.

The plutonium oversaturation experiments did reached steady-state; the three solubilities slowly decreased for the past 100 days, which may have been caused by a restructuring of the solids to more crystalline forms with lower solubilities. Because the concentrations continue to decrease, we are uncertain about how much longer these oversaturation experiments will be run.

We have completed analysis of all samples from the three oversaturation experiments. The solubility experiment at pH 6 has maintained steady-state for more than 100 days. During this period, the solubility experiment at pH 8.5 dropped by a factor of two, to the level of the pH 6 experiment. Another assay will be taken before we stop these experiments and begin to perform oxidation-state determinations.

A faulty rubber O-ring created problems with the Am/Nd experiment at pH 7. Because the experiments were run at 60°C, all rubber seals in the experimental cells were renewed and leak-tested before starting the oversaturation experiments. In spite of this, the solution in the pH 7 experiment evaporated from the cell by the seventh assaying, which did not happen in any other experiments. Because of this, the oversaturation experiments were temporarily stopped, and some of the solid was set aside for x-ray diffraction. The seventh assay consisted of filtering the last aqueous volume from the cell for gamma-pulse height analysis. The concentration determined, 10^{-7} M, was two orders of magnitude higher than the previous two, and probably not valid because the filter may have allowed some precipitate to pass through or the assay may have been contaminated during preparation. The undersaturation experiment was started after all rubber O-rings were changed again, and after twenty days, no detectable evaporation occurred. Because results from the Np undersaturation experiments are reproducing the results from oversaturation, this Am/Nd undersaturation experiment should provide us with the solubility of Am/Nd in UE-25 p#1 water at 60°C. (The oversaturation experiment will be repeated if necessary.)

Regarding the LBL QA effort, work continues on the scientific technical procedure "Concentration Determination of Soluble Radionuclides from Data Provided by a Low-Energy Gamma Counting System." It will be submitted for review next month.

Planned Activities

Efforts in all above mentioned areas will continue.

Continued on next page

Problem Areas None

Milestone Progress

3010
30 June 1991
Report on Measured Solubilities of Pu, Am, and Np in J-13 Groundwater from Oversaturation Conditions
Submitted 7/29/91
In revision.

3031
30 September 1992
Plutonium(IV) and Plutonium(VI) Carbonate Speciation Studies by NMR and PAS Spectroscopies
On schedule.

3329
30 September 1992
Report on Neptunium, Plutonium, and Americium Solubility Experiments in UE-25 p#1 Water from Oversaturation.
On schedule.

3330
1 January 1993
Evaluation of Alternative Detection Schemes in Photoacoustic Spectroscopy
Early completion anticipated.

Publications

D. L. Clark, D. E. Hobart, P. D. Palmer, J. C. Sullivan, and B. E. Stout
Carbon-13 NMR Characterization of Plutonyl(VI) Aqueous Carbonate Complexes
Journal article, *Journal of the American Chemical Society*
In preparation.

D. L. Clark, C. D. Tait, D. E. Morris, D. E. Hobart, S. A. Ekberg, and P. D. *Palmer*
Plutonium(IV) and Plutonium(VI) Carbonate Speciation Studies by NMR and PAS Spectroscopies
LA-series report
In preparation.

D. L. Clark, J. G. Watkins, D. E. Morris, and J. M. Berg
Molecular Models for Actinide Speciation
LA-series report
In preparation.

L. E. Hersman, P. D. Palmer, and D. E. Hobart,
Preliminary Evidence of a Siderophore/Plutonium Complex
Journal article, *Journal of Applied and Environmental Microbiology*
In preparation.

D. E. Hobart, D. L. Clark, P. D. Palmer, J. C. Sullivan, and B. E. Stout
Carbon-13 NMR Characterization of Americyl(VI) Aqueous Carbonate Complexes
Journal article, *Inorganic Chemistry*
In preparation.

Continued on next page

D. E. Morris and D. L. Clark
Spectroscopic Studies of the Hydrolysis of UCl_4: Spectral Effects of Ligand Exchange
LA-series report.
In preparation.

D. Morris, *et al.*
Speciation of Plutonium in Carbonate Media
Conference abstract, *Materials Research Society*
In preparation.

H. Nitsche, R. C. Gatti, E. M. Standifer, S. C. Lee A. Miller, T. Prussin,
R. S. Deinhammer, H. Maurer, K. Becraft, S. Leung, and S. A. Carpenter
Measured Solubilities and Speciations of Neptunium, Plutonium, and Americium in a Typical Groundwater (J-13) from the Yucca Mountain Region
LA-series report
In revision.

C. D. Tait, D. E. Morris, J. M. Berg and W. H. Woodruff
Evaluation of Alternative Detection Schemes in Photoacoustic Spectroscopy
Journal article, *Analytical Chemistry* or *Reviews of Scientific Instrumentation*
In preparation.

C. D. Tait, D. E. Morris, S. A. Ekberg, P. D. Palmer, and J. M. Berg
Plutonium Carbonate Speciation Changes with pH
Conference abstract, *American Chemical Society National Meeting Program*, April 1992
Approved by YMPO.

C. D. Tait, *et al.*
Plutonium Carbonate Speciation Changes
Journal article, *Inorganic Chemistry*
In preparation.

WBS 1.2.3.4.1.4 Radionuclide Retardation by Dispersive, Diffusive, and Advective Processes

Objective

The objectives of this task are to determine the rate of radionuclide movement along the potential flow paths to the accessible environment and to examine the effect of diffusion, adsorption, dispersion, anion exclusion, sorption kinetics, and colloid movements in the flow geometries and hydrologic conditions expected to exist along the flow path to the accessible environment in the scenarios used for perform assessment.

Activities and Accomplishments

This month we continued neptunium (Np) transport work using crushed-tuff columns made from tuff G4-1530.3. Last month we reported that only 35% of the Np injected in the column was recovered in the elutions, corresponds to a K_d ~3 ml/g, and consequently we observe retardation of Np.

This month we divided the column into eighteen 4-inch sections. The dry weight of each section is given in Table 1.

Table 1

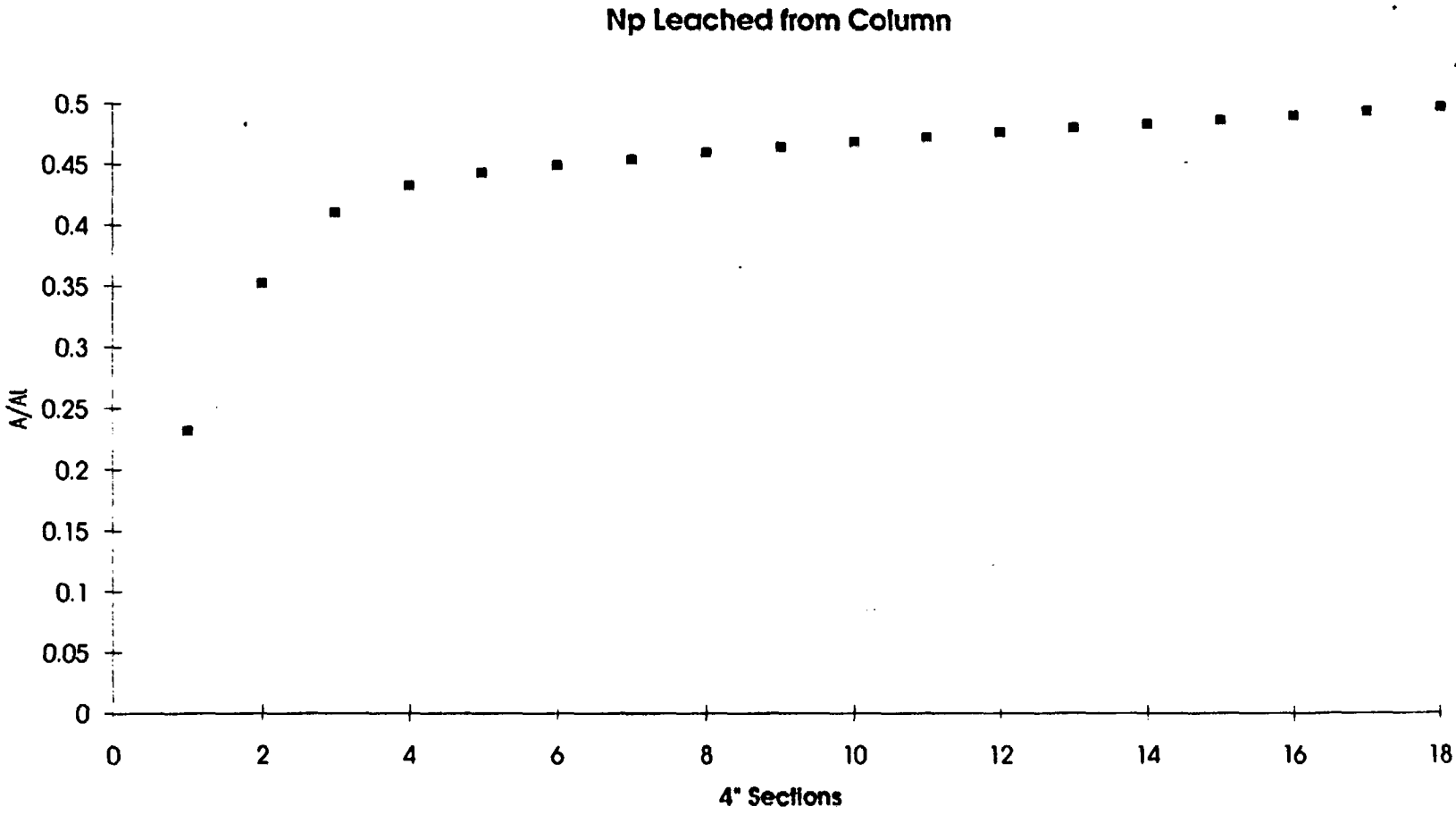
Section	Dry Weight (in grams)
1	2.0
2	2.0
3	2.5
4	2.0
5	1.8
6	1.8
7	1.7
8	1.8
9	1.9
10	1.7
11	1.7
12	1.6
13	1.6
14	1.7
15	1.5
16	1.6
17	1.6
18	1.3

We leached the Np from the tuff section, and 50% of the Np injected into the column was found in the leachate. Consequently, we can account for 85% of the Np injected into the column. The exploration for the unaccounted for 15% is that the leaching procedure could not extract 100% of the NP the tuff. As indicated in Figure 1, 40% of the NP was sorbed onto the tuff in Sections 1-3.

To support our transport experiments we continued Np batch-sorption experiments using tuffs G4-1530 and G4-275 and USWH-3, UE-25 p#1, and J-13 waters. In addition, we conducted batch-sorption experiments using Np solutions and pure mineral separates found in the following tuffs: hematite, montmorillonite, clinoptilolite, and quartz. Figure 1 shows the relative cumulative activity of Np found in the solid phase of the column sections.

Continued on next page

Figure 1


Np Leached from Column
A/Al
0.5
0.45
0.4
0.35
0.3
0.25
0.2
0.15
0.1
0.05
0
0
2
4
6
8
10
12
14
16
18
4" Sections

Staff filled out six technical data information forms and designed parameter screens for the dynamic transport and diffusion tasks.

Staff began to organize the colloid workshop that will be held in early 1993.

A. Mitchell served as briefer in the Yucca Mountain Site Characterization Project Open House.

Planned Activities Continue work described above.

Problem Areas None

Milestone Progress

3040
30 September 1992
Kinetics of Sorption on Columns of Pure Minerals

3044
31 August 1992
Letter Report on Assessment of Available Techniques for Unsaturated Column Transport Experiments
In preparation.

3027
31 March 1992
Report on Sorption by Batch and Column Techniques

Publications

I. R. Triay
Radionuclide Migration in Tuff under Diffusive Conditions
Conference Paper, *Proceedings of the Migration '91, Jerez de la Frontera, Spain, 14–18 October 1991*
In preparation.

I. R. Triay, A. J. Mitchell, and M. A. Ott
Radionuclide Migration Studies for Validating Sorption Data—Past, Present, and Future
Conference paper, *Proceedings of the DOE/Yucca Mountain Site Characterization Project Radionuclide Adsorption Workshop at Los Alamos National Laboratory September 11-12, 1990. (LA-12323-C, 1992)*
Revision submitted to YMPO.

WBS 1.2.3.4.1.5.1 Retardation Sensitivity Analysis

Objective

The objectives of this task are to construct a geochemical/geophysical model of Yucca Mountain and to use this model to examine the physical and chemical controls on radionuclide transport along flow paths to the assessable environment.

Activities and Accomplishments

QA and Programmatic. Certification of TRACRN continued. The Design Phase Baseline was reviewed by the TRACRN review committee, and RIDS were informally generated. The Software Design Document, the Models and Methods Summary, and some software prologs were modified in response to reviewer comments. The Verification and Validation (V&V) Plan Baseline was completed and submitted for review.

Verification also continued. Comparisons between the two codes, TRACRN and FEHMN, were being run for unsaturated-zone flow (water only and air/water) problems and for transport problems. Comparison of the codes for saturated flow was good and was reported in the May monthly report.

The user's manual is nearly complete.

Certification of FEHMN and GZSOLVE continued. G. Zyvoloski worked with B. Robinson and Z. Dash of Task 1.2.3.3.1.3.1 on documentation and verification of these software applications.

Planned Activities

Continue certification of TRACRN.

Problem Areas

None

Milestone Progress

3052
31 July 1992
Baseline Documentation for TRACRN

Publications

K. Birdsell, K. Eggert, and B. Travis
Three-Dimensional Simulations of Radionuclide Transport at Yucca Mountain
Journal article, *Radioactive Waste Management and The Nuclear Fuel Cycle - Special issue on the Yucca Mountain Project*
Revised version submitted to YMPO.

K. Birdsell, K. Campbell, K. Eggert, and B. Travis
Sensitivity Analysis of Integrated Radionuclide Transport Based on a Three-dimensional Geochemical/Geophysical Model
Conference proceedings, *Proceedings of the DOE/Yucca Mountain Site Characterization Project Radionuclide Adsorption Workshop at Los Alamos National Laboratory September 11-12, 1990.* (LA-12323-C, 1992)
In press.

WBS 1.2.3.4.1.5.2 Demonstration of Applicability of Laboratory Data

Objective

The purpose of this study is to design and conduct experiments to evaluate the applicability of laboratory data and to test models used in the Radionuclide Transport Program to determine far field radionuclide transport. Both intermediate- and field-scale experiments and natural analogues will be assessed for their potential to provide the required data.

Activities and Accomplishments

Mission 2001 cost and schedule planning for both Lawrence Berkeley Laboratory (LBL) and Los Alamos for this study were completed. A meeting was held in Las Vegas on 8 June to discuss LBL input for Mission 2001 with DOE and M&O staff. (LBL input was submitted to the M&O because the workstation at Los Alamos cannot accept input from other participant.)

E. Springer attended the joint Geochemistry Integration Team / Hydrology Integration Team meeting held in Denver during June 9-11.

This task was the subject of an internal audit.

International Programs: Natural Analogs. Input was provided to the DOE for the "International Programs Annual Report." See Appendix for this information.

Planned Activities

Continue to develop study plan.

Problem Areas

None

Milestone Progress

No FY91 milestones.

Publications

C. Loeven
A Summary and Discussion of Hydrologic Data from the Calico Hills Nonwelded Hydrogeologic Unit at Yucca Mountain, Nevada (LA-12376-MS, 1992)
LA-series report
Approved by YMPO.
Final editing before publication.

WBS 1.2.5 Regulatory and Institutional

Objective

The purpose of this task is to coordinate the regulatory and institutional Project requirements within the Los Alamos programmatic structure. The focus of this coordination effort is on the integration of the technical work within the regulatory and institutional framework.

Management and Integration

Significant effort was made to put forth changes to the SCP Baseline. In particular, the rock-varnish work to support volcanism, erosion, and neotectonic studies was identified discretely. PACs changes were initiated. Work began to evaluate rock-varnish data in support of the erosion issue resolution.

Study Plans

Water Movement Test, R1 (8.3.1.2.2.2). R1 has been approved by DOE but not by NRC. A revision incorporating NRC and State of Nevada comments was submitted to the YMPO on 17 October 1991.

Diffusion Test in the Exploratory Studies Facility, R0 (8.3.1.2.2.5). This study plan was approved by the YMPO and DOE/HQ in May 1992.

Testing of the C-Hole Sites With Reactive Tracers, R1 (8.3.1.2.3.1.7). In February 1990 DOE/HQ issued this study plan as a controlled document; it was then sent to the NRC for comments. In January 1992 we were requested by DOE to revise NRC comments. The revision is in progress.

Ground Water Chemistry Modeling, R0 (8.3.1.3.1.1). In March 1991 this study plan was submitted to the project office for review .

Mineralogy, Petrology, and Chemistry of Transport Pathways, R3 (8.3.1.3.2.1). In August 1990 the NRC approved the study plan. In October 1991 we were asked to revise the study plan; in January 1992 we submitted revised comments to T. Bjerstedt.

History of Mineralogy and Geochemical Alteration at Yucca Mountain, R0 (8.3.1.3.2.2). The Project Office approved the study plan in December 1991 and submitted it to the NRC in January 1992 for comments.

Natural Analogue Hydrothermal System in Tuff (8.3.1.3.3.1). This is an out-year activity.

Kinetics and Thermodynamics of Mineral Evolution and Conceptual Model of Mineral Evolution, R0 (8.3.1.3.3.2; 8.3.1.3.3.3). A comment resolution meeting for DOE/HQ and Project Office comments was held in March 1990; revision on this activity has been deferred because funds have not been allocated.

Sorption Studies and Sorption Modeling, R0 (8.3.1.3.4.1; 8.3.1.3.4.3). A new study plan has been issued for internal review.

Biological Sorption and Transport, R1 (8.3.1.3.4.2). Revisions incorporating DOE/HQ and Project Office comments were submitted in May 1991. Additional revised text was submitted in August 1991.

Continued on next page

Dissolved Species Concentration Limits, and Colloid Formation and Stability, R0 (8.3.1.3.5.1; 8.3.1.3.5.2). In November 1990 the project office submitted comments to Los Alamos to revise; that revision is in progress.

Dynamic Transport Column Experiments, R0 (8.3.1.3.6.1). A comment resolution meeting for DOE/HQ and Project Office comments was held in August 1990; revisions are in progress and are expected to be completed by June 1992.

Diffusion, R0 (8.3.1.3.6.2). A comment resolution meeting for DOE/HQ and Project Office comments was held in August 1990; revisions are in progress and are expected to be completed by June 1992.

Retardation Sensitivity Analysis, R0 (8.3.1.3.7.1). A revision incorporating DOE/HQ and Project Office comments was submitted in June 1991. In October additional comments were received from SAIC, P. Cloke. The comments have been addressed and were submitted to the P. Cloke in March 1992.

Demonstration of the Applicability of Laboratory Data to Repository Transport Calculations, R0 (8.3.1.3.7.2). This study plan is in preparation.

Gaseous Radionuclide Transport Calculations and Measurements, (8.3.1.3.8.1). Funds have not been allocated.

Probability of Magmatic Disruption of the Repository, R0 (8.3.1.8.1.1). This study plan was approved by the Project Office in September 1990 and by the NRC in October 1991.

Physical Processes of Magmatism and Effects on the Potential Repository, R0 (8.3.1.8.1.2). Study plan is complete and has been issued for internal review.

Characterization of Volcanic Features, R0 (8.3.1.8.5.1). Accepted by NRC in September 1990.

WBS 1.2.6 Exploratory Studies Facility

Objective

These Exploratory Studies Facility (ESF) tasks address the issues and information needs associated with the ES-based characterization of Yucca Mountain to determine the suitability of permanently isolating high-level nuclear waste from biosphere in a geologic repository.

Activities and Accomplishments

Continue work to develop a Tracers, Fluids, and Materials (TFM) information program consistent with the plan issued by the Project (YM 91-23) and prepared by Los Alamos. Support civilian Radioactive Waste Management Systems Management and Operating (CRWMS M&O) contractor efforts on developing a position paper for prototyping needs. Developed input for the position paper and participated in meetings. Prepared briefings for bi-weekly ESF ED&D meetings. Participated in ESF excavation and ESF testing planning meeting. Reviewed proposals submitted by Raytheon Services Nevada (RSN) and CRWMS M&O in support of Integrated Data System (IDS) design services. Provided informal comments to Engineering and Development Division (ED&D) on the proposals.

Planned Activities

Continue to participate in Test Integration Group (TIG) meetings.

Continue to develop definitive design-related information for tests to be performed in the launch chamber.

Continue to support integration meeting such as ESF design, TIG, Sample Management Facility (SMF) surface-based testing and its interface with ESF testing.

Support ED&D effort in developing rationale and justification for the need to have a prototype test facility for Yucca Mountain.

Support ESF design review, management, and technical.

Develop interfaces for testing and the ESF design.

Revise and update PSAR as required.

Start identifying Project IDS planning.

Initiate work to develop new networks for ESF testing.

Prepare test planning packages for launch chamber tests.

Continue with gathering TFM information from participants.

Start developing data files for TFM.

Prepare Title II Test Planning Packages.

Continue with ESF sample and test coordination efforts.

Continued on next page

	Support RSED Director as requested.
Problem Areas	None
Milestone Progress	None

WBS 1.2.6.8.4 Integrated Data System

Objective

The integrated data system (IDS) supports the Exploratory Studies Facility (ESF) test program by providing a central facility to automatically measure and control aspects of the ESF tests. The primary purposes of the IDS are to assist the principal investigators (PI's) in acquiring high-quality test data in a uniform, controlled fashion and to transfer those data to the PI's organizations for data management and analysis.

Activities and Accomplishments

This activity has been deferred.

WBS 1.2.9.1.2.4 Technical Software Management

Objective

The purpose of this activity is to manage the development, implementation, and use of all software employed on activities that will support a license application; to manage the configurations of all software and computational data; and to provide tools and procedures that support these activities. Technical software management continued to perform the quality assurance, configuration management, and engineering tasks that are required by the Los Alamos Software Quality Assurance Plan for the Yucca Mountain Project.

Activities and Accomplishments

This activity has been deferred because of lack of funds.

WBS 1.2.9.1.4 Records Management

Objective

The objective of this task is to manage records and documents related to the licensing of a geologic repository for the disposal of high-level radioactive waste by developing, implementing, and maintaining a comprehensive, automated, and integrated information management system.

Activities and Accomplishments

The Records Processing Center (RPC) received 94 records and records packages and rejected 19, which were returned to the originators for corrections.

The Central Records Facility (CRF) returned 7 records or records packages for corrections. The RPC resubmitted 22 corrected records to the CRF.

The RPC was the subject of an internal audit.

WBS 1.2.9.3 Quality Assurance

Objective

The Quality Assurance (QA) Program supports Los Alamos Yucca Mountain Site Characterization Project participants and ensures that their efforts provide data and evidence admissible for the repository-licensing process.

Activities and Accomplishments

Grading Reports. Los Alamos has thirty-three approved grading reports. Six grading reports (three are revisions of existing reports) are in review at the Project Office.

Records/Document Control. Seven detailed technical procedures (DPs) were deleted. DP 130, R0 (Geologic Sample Preparation) and DP 90, R1 (Measurement of Moisture Content in Soil Samples) were issued. Four quality administrative procedures (QPs) were also approved: QP 12.1, R6 (Measuring and Test Equipment); QP 02.11, R0 (Personnel Orientation); QP 3.24, R0 (Submittal of Design and Test Related Information); and QP 3.25, R0 (Review of Design and Test Related Information).

Program Development. Eighteen QPs are in various stages of revision. Fifteen members of the quality group reviewed the QARD.

Audits. Audit LANL-AR-92-07 was extended to 7-6-92. The audit report for EES-5 activities (LANL-AR-92-06) was approved and issued. The annual management assessment report of the Los Alamos QA program is in final preparation.

Planned Activities

DOE will conduct audit YMP-92-19. QP revisions will continue. A CCB meeting will be convened in mid July.

Problem Areas

It will be necessary to hire a new configuration manager and software support personnel. This may cause a delay in processing of some SCRs. Revision of the SQAP and associated procedures will be postponed until budget and personnel issues can be addressed.

Publications

S. L. Bolivar
The Los Alamos National Laboratory Yucca Mountain Site Characterization Project Quality Program, A Progress Report for January 1, 1990 - December 31, 1991.
In internal review.

S. L. Bolivar and J. L. Day
The Role of the Los Alamos National Laboratory Quality Assurance Liaison for the Yucca Mountain Site Characterization Project
Conference paper, ASQC Energy Division Annual Meeting
In preparation.

APPENDIX

ATTACHMENTS AND LEVEL III MILESTONE REPORTS

Radionuclide Retention in the Cigar Lake Spent Fuel Analogue

David B. Curtis, June Fabryka-Martin, Fred Roensch,
Ruben Aguilar, and Moses Attrep
Isotope & Nuclear Chemistry Division
Los Alamos National Laboratory
Los Alamos, New Mexico, USA

1. Research objectives and approach

Radionuclides are produced in the solid earth by naturally occurring nuclear reactions. Many of these nucleogenic nuclides are the same as those that produce the long-term risk associated with the storage of radioactive wastes from nuclear power plants. In nature, ^{99}Tc and ^{129}I are the products of two fission processes, spontaneous fission of ^{238}U, and neutron induced fission of ^{235}U. Plutonium-239 is produced by neutron capture by ^{238}U. If the system in which they are produced is undisturbed on the time-scale of their radioactive half-lives, these nuclides have the simplest of geochemical histories: They are formed and disappear within the phase where the parent element resides. In such systems, the relative proportions of the parent and daughter nuclides are a function of the relative rates of production and decay of the radioactive daughter. In principle, these proportions can be predicted from an understanding of the systematics of the nuclear processes. Comparisons between predicted and actual abundances provide a means of characterizing complex geochemical histories for the radionuclides. Deviations from the predicted abundances manifest differential rates of release, transport, and retention of the daughter radionuclides and their parent elements during windows of time extending back for a hundred thousand to several million years from the present. We are characterizing the retentive properties of uranium-bearing rocks and minerals from a variety of geochemical settings. The collection and correlation of these observations will be used to develop an understanding of processes controlling the release and retention of radionuclides from spent fuel analogues to determine if this understanding is consistent with the conceptual basis for models of radionuclide release being used for performance assessments of geologic repositories for spent nuclear fuel.

2. Cigar Lake - The spent fuel analogue

Uranium in the Cigar Lake deposit is contained primarily in reduced uranium oxides (uraninite/pitchblende) and secondarily in the silicate mineral coffinite (1,2). Primary uraninite was formed 1.3 x 10^9 yr ago (3). It has been severely altered by dissolution, loss of lead, and coffinization . The majority of these processes were associated with the formation of a clay halo, 1.1-1.2 x 10^9 years ago. The presence of sulfide minerals indicates that the ancient alteration processes occurred under reducing conditions (2). Groundwater compositions suggest the current system is also reducing. It is suggested that reducing conditions controlled the water-rock interaction in the uraniferous rock for much of its geologic history (4). Under such conditions, solubility of the uraniferous phases severely limits the concentration of uranium in the groundwater, thereby limiting the release and redistribution of uranium in the solid phase. It appears that uranium-bearing minerals at Cigar Lake have been chemically and physically stable for nearly 10^9 yr. The relative abundances of the nucleogenic radionuclides ^{99}Tc, ^{129}I, and ^{239}Pu and their parent element, uranium, are expected to be in secular equilibrium, reflecting their quantitative retention for the periods of time required to

Preliminary Data—Do Not Reference

establish secular equilibrium (10^5 to 10^7 yr).

From the perspective of this work, the Cigar Lake analogue addresses our understanding of the ability of reduced uranium oxides and their alteration phases to contain radionuclides in a natural setting over a geologic time-scale.

3. Radionuclides in Cigar Lake samples

3.1 Plutonium-239

Abundances of ^{239}Pu have been determined in three samples of Cigar Lake ore by isotope dilution mass spectrometry (5,6). Masses of indigenous plutonium in the analyzed samples are extremely small, and thus the results are vulnerable to inaccuracies from contamination introduced during storage, sampling, or preparation for analysis. Consequently, numerous blanks, samples containing no indigenous plutonium, were periodically analyzed in conjunction with the samples. The results of the analyses of blanks show that measurable plutonium is introduced randomly during analyses. The average quantity of contaminant ^{239}Pu is $8 \pm 6 \times 10^7$ atoms. All samples were corrected for this quantity. The detection limit for ^{239}Pu, defined as three standard deviations above the average measured blank, is 2.4×10^8 atoms. Blank corrected abundances in Cigar Lake samples are given in Table 1.

Table 1. Radionuclide abundances in Cigar Lake samples

Sample	U (%)	$^{239}Pu/U$ ($\times 10^{12}$)	$^{129}I/U$ ($\times 10^{12}$)	$^{99}Tc/U$ ($\times 10^{12}$)	$^{36}Cl/Cl$* ($\times 10^{11}$)
CS 235L	55.6	1.9 ± 0.1	4.85 ± 0.48		2.0
			2.35 ± 0.20		
			1.72 ± 0.13		
			2.10 ± 0.17		
W83A	39.0	3.2 ± 0.2	3.49 ± 0.36	<1.8	4.6
			2.05 ± 0.19		
W83C	46.8	2.8 ± 0.2	4.52 ± 0.44		
			2.75 ± 0.25		

*Unpublished data from Jack Cornett. These analyses were done on a different aliquot than that used for the U, ^{99}Tc, ^{129}I, and ^{239}Pu analyses.

3.2 Iodine-129

Iodine-129 measurements were made by accelerator mass spectrometry. For this purpose iodine was extracted from the rock samples and chemically purified. Initially the element was extracted from a solution produced by sodium peroxide fusion and acid dissolution. However, blank samples analyzed by this method indicated that variable, and often significant quantities of ^{129}I were introduced during analysis. A procedure using high pressure - high temperature acid dissolution in a microwave oven produced insignificant blanks. However, the procedure did not completely dissolve the samples, resulting in uncertainties as to whether the ^{129}I had been quantitatively released from the solid phase. Measured ^{129}I abundances in the Cigar Lake samples are presented in Table 1. These results have not been corrected for blanks.

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Until the constraints on the analysis for this nuclide are better understood, these results should be viewed as limiting the actual concentration of indigenous ^{129}I in the sample.

3.3 Technetium-99

One of the samples was analyzed to measure the concentration of ^{99}Tc. Blank samples showed that spurious large quantities of contaminant ^{99}Tc were introduced into samples during the analyses. Consequently, this single result was not corrected for blank and is reported as an upper limit in Table 1.

3.4 Uranium

Abundances of uranium in the samples was measured by alpha-spectrometry at the AECL Whiteshell Laboratory. The results are included in Table 1.

4.0 Models of radionuclide production

In a static system, the abundances of the products of spontaneous fission, ^{99}Tc and ^{129}I, are in direct proportion to the uranium abundance. The quantities of the products of neutron-induced fission and neutron capture are a function of the neutronics - the neutron flux and energy spectrum in the rock. A continuous energy neutron/photon transport code was used to predict neutron capture produced radionuclide abundances in samples from unaltered systems. The code, called the Monte Carlo Neutron and Photon (MCNP) transport code was developed at Los Alamos during the Manhattan Project, and has been updated and maintained since then. The user specifies the geometry of the problem, the composition of the materials, evaluations of the nuclear reaction cross-sections, and the characteristics of the neutron sources. The model output tallies the events per starting neutron as a function of the energy of the captured neutron; For example the relative proportions of ^{235}U fission induced by fast, epithermal, and thermal neutrons. The MCNP predictions of events per initial neutron are converted to events per unit mass per unit time by multiplying by the measured neutron production rate per unit mass of sample. An overview of the modelling process is shown in Figure 1.

4.1 Models of infinite homogeneity

The first applications of the MCNP for these purposes were done by measuring the critical compositional parameters in gram-sized samples and assuming that these were representative of the geologic environment from which the samples were taken. This approach essentially assumes that the environment from which the sample was taken was infinitely homogenous. A comparison between the $^{239}Pu/U$ predicted by such models and the measured ratios in samples from Cigar Lake and other unaltered uranium deposits are shown in Figure 2. Of the eleven samples, the predicted and measured values compare favorably in only one. Subsequent modeling demonstrated that the neutronics in a gram-sized sample is a function of the quantity and distribution of selected elements within the 0.125 cubic meters of rock that encompassed the sample in its natural setting. We conclude that in heterogeneous media, such as an ore body, it is difficult to reconstruct the quantity and distribution of critical elements with sufficient accuracy to make accurate predictions of the abundances of neutron produced radionuclides (7).

4.2 Correlated abundances of nucleogenic radionuclides

Without sufficient information to accurately predict the unaltered abundances of nucleogenic nuclides, we investigated the possibility of characterizing their *in-situ* retention

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by comparing measured and predicted *correlations* between the radionuclides. Figure 3 shows such a correlation between $^{129}I/U$ and $^{36}Cl/Cl$. Chlorine-36 is a radionuclide with a half-life of 3×10^5 yr, produced by neutron capture by the stable chlorine isotope ^{35}Cl. In uranium ores, the ratio is a measure of the neutron fluence over about the last million years, or the period of time that chlorine has resided in the sample, whichever is the shortest period. It is subject to perturbation only by the addition of chlorine from another source. The shaded area within the graph in Figure 3 represents the predicted range of values for the two ratios produced by varying the compositional parameters in the radionuclide production model to mimic the range of compositions in the Cigar Lake ore. The narrow band demonstrates a similarity in the dependence of the two production reactions on the neutronics. Data from the Cigar Lake samples are plotted on the diagram. Although the large uncertainties in the measured abundances of ^{129}I (section 3.2) result in considerable ambiguity in the interpretation of these data, they are however consistent with the anticipated quantitative retention of the iodine fission product in Cigar Lake samples. We have included the results of an analyses of an altered sample from the weathered zone of the Koongarra deposit. This result demonstrates a clear case of the failure to retain ^{129}I *in-situ*. If the uncertainties in the analytical data can be reduced, this comparative technique may be an effective method of characterizing the degree of retention of ^{129}I.

Figure 4 is a similar diagram showing the predicted correlations between $^{129}I/U$ and $^{239}Pu/U$. It further illustrates the difficulty in predicting the unaltered abundance of ^{239}Pu. The polygons in the figure represent the predicted range of ratios produced by varying the abundances of the heavy rare earth elements and hydrogen, elements which have the most profound effect on the neutronics, within the measured composition of the samples. The failure of any sample to fall within its predicted range is again a demonstration of the failure of the assumption of infinite homogeneity in the MCNP calculations. All three of the samples fall close to the region described by assuming that the materials controlling the neutronics had the composition of average ore; again consistent with our expectations of quantitative retention by the uranium minerals in the deposit. However, unlike the narrowly defined correlation between $^{36}Cl/Cl$ and $^{129}I/U$ shown in Figure 3, the dependence of the ^{129}I and ^{239}Pu production processes are not sufficiently similar to give tight correlations, and the procedure is not very definitive for characterizing the relative retention of the two radionuclides.

4.3 Spatial distribution of nucleogenic radionuclides

Because of the large volume of rock controlling the neutronics at a point, spatial variations of the abundances of radionuclides produced *in-situ* by neutron reactions should be attenuated relative to the scale of compositional variations in the ore. Hence, we believe that we can reduce interpretative ambiguity by characterizing the distribution of nucleogenic radionuclides in samples with well-characterized spatial relationships. For this purpose we have selected samples from Cigar Lake core 220 (**Reference**)for analysis. The core includes 6.3 meters of sample from the ore zone. The section of ore is compositionally highly variable. Uranium concentrations vary from 45% to less than 0.1 %. The lithology includes rock containing massive uranium, massive clay, and sedimentary rock described as sandy rubble. We have done preliminary modelling of the parent/daughter ratios of the nucleogenic radionuclides through the section of core. Compositional variations were artificially constructed by assuming the samples to be a two component mixture of uraniferous material and

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non-uraniferous material. The composition of the two components was reconstructed from our analyses of sample CS235L. *This is completely artificial*, and intended only to illustrate the proposed approach. Elemental analyses of the actual samples are being done by AECL. The results of this preliminary modelling are shown in Figure 5. Predicted $^{239}Pu/U$ ratios show much less variability than do the uranium concentrations. With the exception of three spikes, the distribution of relative abundances of the two actinides through the core consists of broad, fairly level peaks centered around the highest concentrations of uranium. The spikes occur in thin strata of low-uranium rock sandwiched between thick sections of high-uranium material. The total range of predicted ratios is 0.3 - 13 x 10^{-12}. Omitting the anomalous spikes, the range is 0.3 - 5 x 10^{-12}, consistent with measured $^{239}Pu/U$ ratios on the three Cigar Lake samples (Table 1).

The spatial distributions of $^{129}I/U$ and $^{36}Cl/Cl$ demonstrate the expected correlation between these two ratios. Peaks in the ratios are associated with highs in the uranium content. The range in the $^{129}I/U$ ratio is not as great as that of the chlorine isotopes, because ^{129}I from spontaneous fission - which is independent of the neutron flux - comprises a significant portion of this radionuclide concentration in the less uraniferous samples. In unaltered samples the ratio $^{129}I/U$ will never be less than the fixed secular equilibrium ratio produced by spontaneous fission.

The predicted spatial variation of $^{99}Tc/U$ is virtually independent of compositional variations. This reflects the fact that essentially all of the ^{99}Tc in the ores is the product of spontaneous fission. This is fortuitous in that it allows us to evaluate the degree of retention of technetium independent of model predictions.

5.0 Conclusions

The work that has been done demonstrates the potential of studies of nucleogenic radionuclides in understanding the processes affecting the long-term retention and loss of radionuclides from reduced uranium oxides and their alteration phases in geologic environments. Such understanding will be critical in validating models of radionuclide release used in spent fuel repository assessments. However, the work also emphasizes what must be done to realize this potential. Blanks associated with the analyses of ^{99}Tc and ^{129}I must be controlled so that the precision of the measurements is sufficient to meaningfully interpret the retention and loss of these two fission products in a variety of geochemical settings. Models of neutron-induced radionuclide production must be developed to allow us to evaluate the degree of retention of these nuclides. Development of an accurate modelling method is critical in understanding the geochemical behaviour of plutonium, less so for ^{129}I, and of no consequence in characterizing the geochemistry of technetium.

We anticipate that the realization of these objectives for samples from Cigar Lake will demonstrate the retentive properties of uranium minerals in a reducing environment, and provide a fiducial reference for evaluating such properties in different geochemical settings.

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Figure Captions

Figure 1. A schematic diagram demonstrating the model used for predicting nuclide production in the solid earth. The figure demonstrates that ^{99}Tc and ^{129}I are produced by spontaneous and neutron induced fission. Plutonium-239 is produced exclusively by neutron capture. The figure also shows the input parameters required the MCNP code.

Figure 2. This graph showing the relationship between measured and predicted $^{239}Pu/U$ values in unaltered ores demonstrates the inaccuracies associated with a model that assumes an infinitely homogenous medium with a composition defined by the sample. Agreement between the actual value and the predicted one would fall on the 45^0 line. With few exceptions, this model over-predicts the relative ^{239}Pu abundance.

Figure 3. The correlation between the ratios $^{129}I/U$ and $^{36}Cl/Cl$ in samples of Cigar Lake ore to fall within the model predicted correlation between these ratio pairs. The agreement between the measured and predicted correlations suggests that these elements have been retained *in situ*. However, the large range of measured $^{129}I/U$ in each sample does not makes the conclusion compelling. Data from the Koongarra uranium deposit is included to demonstrate the lack of agreement between the model predicted and measured correlations in a system in which the elements have not been retained in their host phases.

Figure 4. The failure of any of the data to fall within the predicted regions of correlation between the neutron capture products ^{129}I and ^{239}Pu for individual samples again demonstrates the failure of the model of infinite homogeneity. However, all of the data falls near the correlation predicted assuming production in a media with the composition of average Cigar Lake ore. This agreement between prediction and measurement is again consistent with our assumption that the Cigar Lake minerals have retained the radionuclides.

Figure 5. The figure shows model predictions of the spatial distribution of radionuclides produced in core 220. The distributions are for purposes of demonstration only. They do not reflect reality because the uranium content is the only known compositional parameter, the remainder of the sample compositions were simulated. The model demonstrates the damping of variations in $^{239}Pu/U$ relative to the uranium content, the correllated variations between $^{129}I/U$ and $^{36}Cl/Cl$, and the constancy of $^{99}Tc/U$ independent of uranium content.

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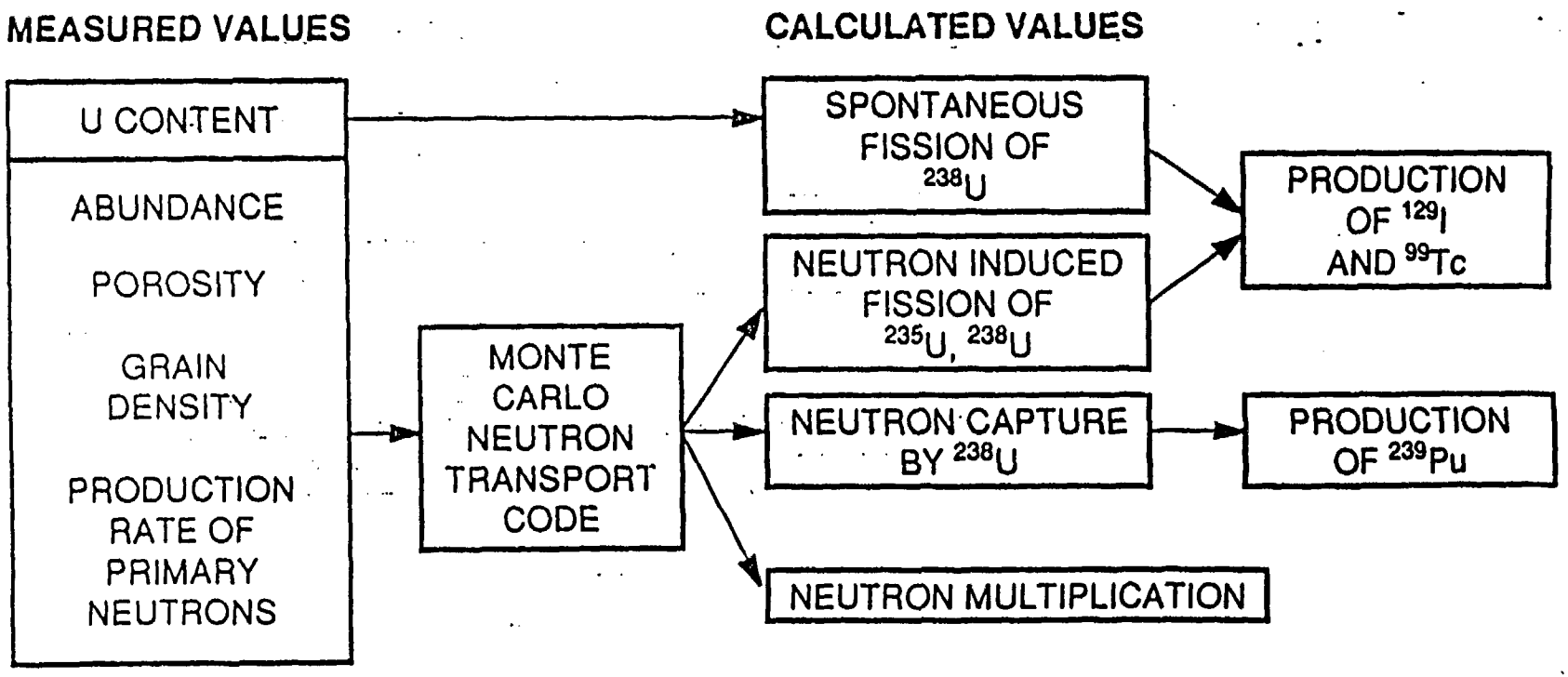
MODEL FOR PREDICTING NUCLIDE PRODUCTION



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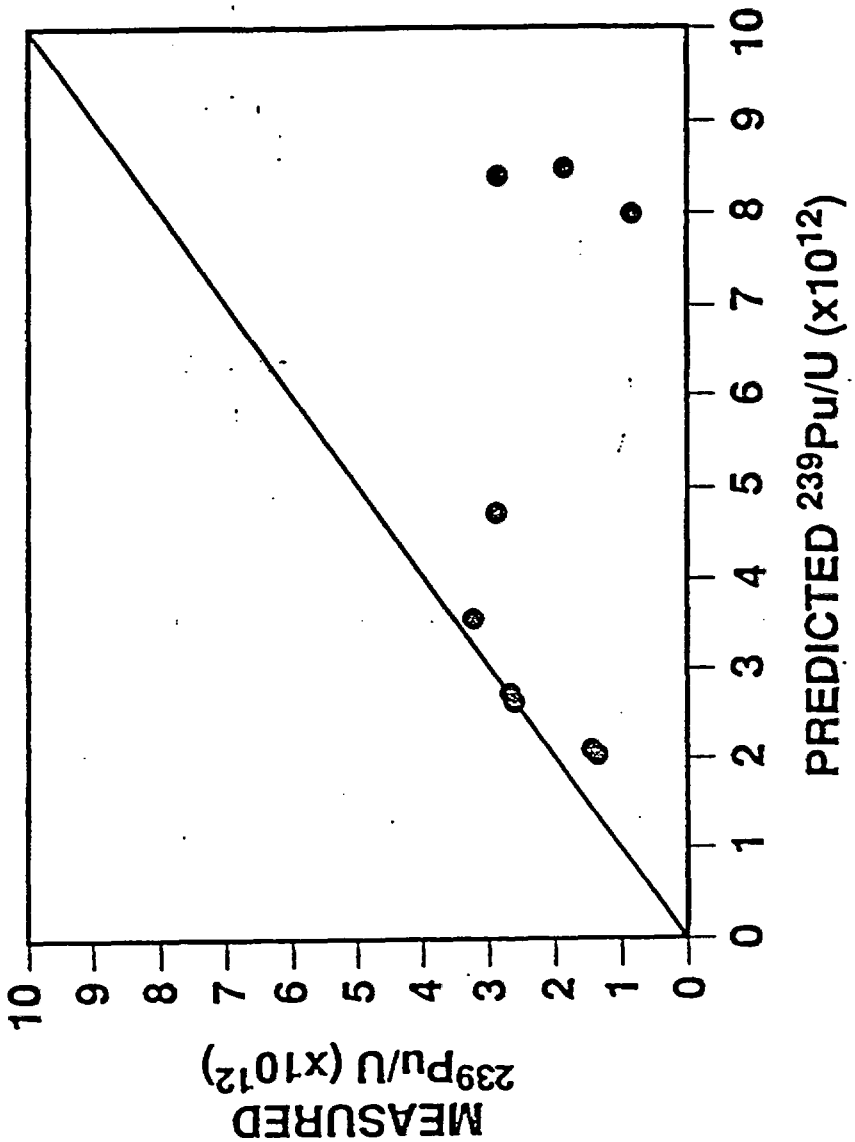

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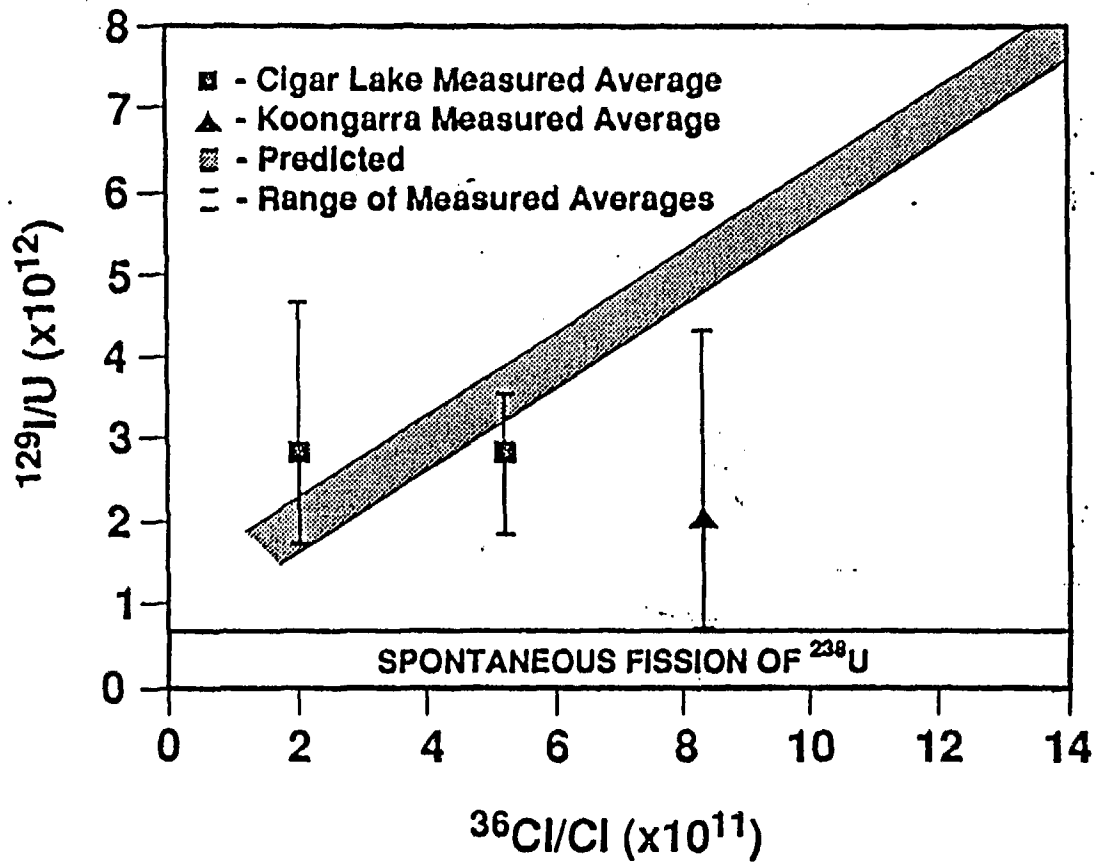

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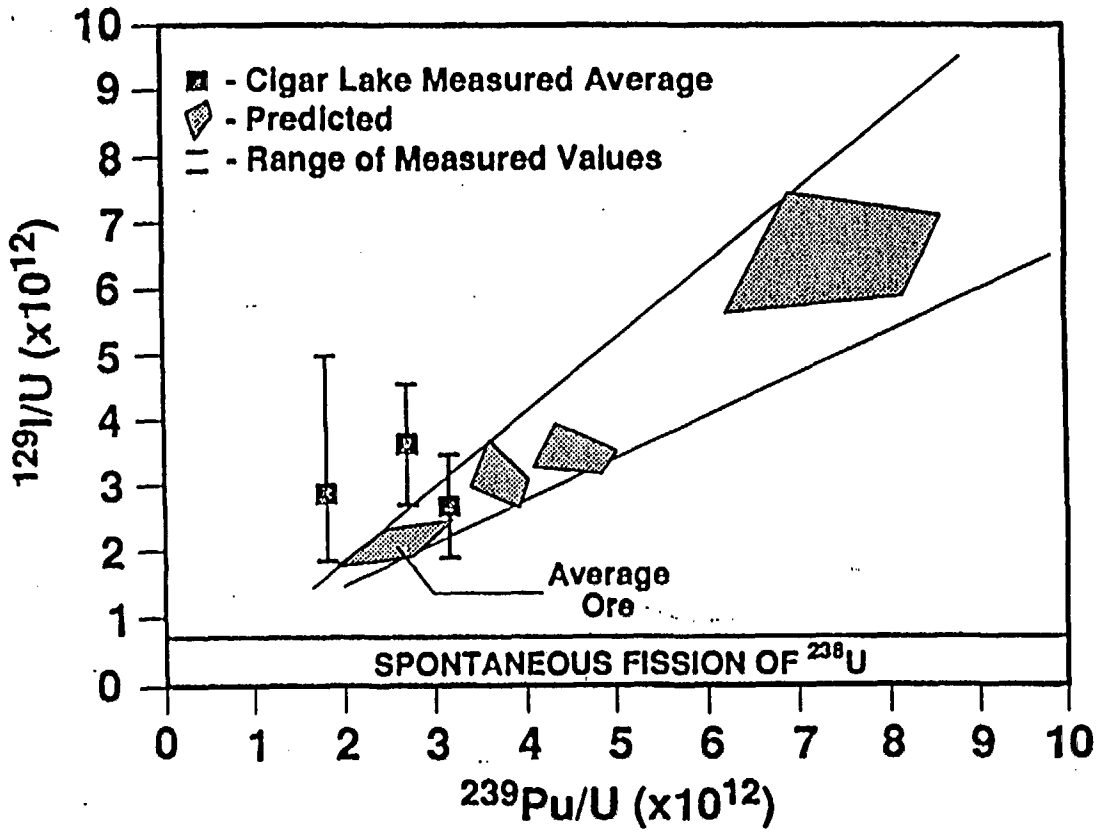

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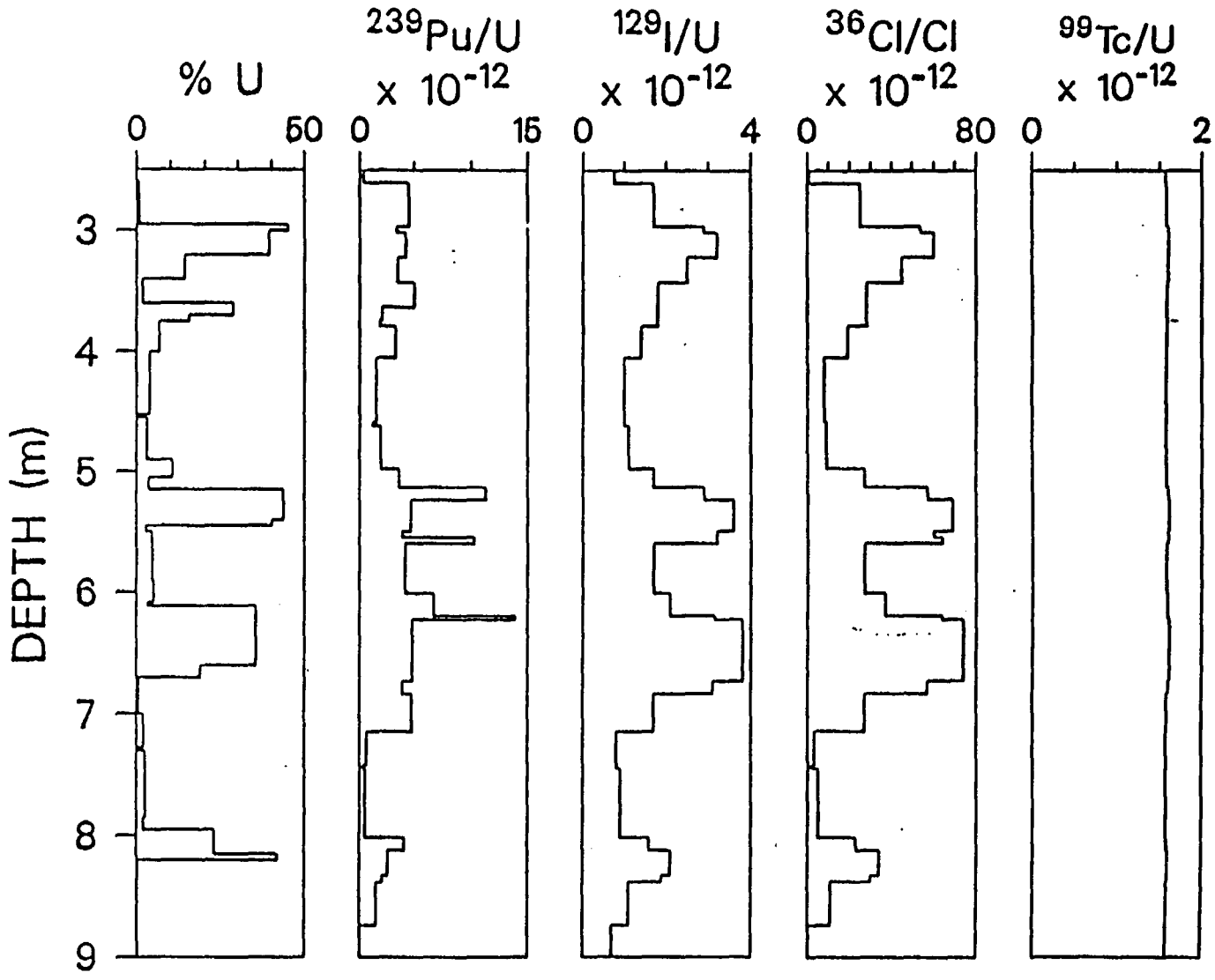
PROFILES OF RADIONUCLIDE PRODUCTION IN CIGAR LAKE HOLE 220



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