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September 23, 1992

TWS-EES-13-09-92-044

Mr. Carl P. Gertz, Project Manager Yucca Mountain Site Characterization Project Office US Department of Energy P.O. Box 98608 Las Vegas, NV 89193-8608

Dear Mr. Gertz:

SUBJECT: LOS ALAMOS MONTHLY ACTIVITY REPORT-AUGUST 1992

Attached is the Los Alamos Monthly Activity Report for August 1992. This internal document describes our technical work in detail; however, the report has not received formal technical or policy review by Los Alamos or the Yucca Mountain Site Characterization Project. Data presented in this document represent work progress, are not referenceable, and are not intended for release from the US Department of Energy. If you have changes to our distribution list, please call me at (505) 667-0916.

Sincerely.

san H. K/c\_ Susan H. Klein

SHK/elm

WASTE

PDR

PDR

WM-11

Attachment: a/s

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S. H. Klein, IS-11, MS J521 CRM-4, MS A150 An Equal Opportunity Employer/Operated by the University of California 212090066 921201

ENCLOSURE 1

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## LOS ALAMOS NATIONAL LABORATORY

## YUCCA MOUNTAIN SITE CHARACTERIZATION PROJECT

## Monthly Activity Report

## August 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
 Entered data into the Automated Technical Data Tracking System for Activities 8.3.1.3.2.1.2, "Calcite Deposits In Drill Cores USW G-2 and USW GI-3/G-3 at Yucca Mountain, Nevada"; 8.3.1.3.2.2.1, "Pedogenesis of Siliceous Calcretes of Yucca Mountain, Nevada"; 8.3.1.3.2.2.2, "Dehydration and Rehydration of a Tuff Vitrophyre"; and 8.3.1.3.2.2.1, "Bedrock Breccias Along Fault Zones Near Yucca Mountain, Nevada."
 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.
 Caisson Experiment (WBS 1.2.1.4.6)

Activities and<br/>AccomplishmentsThe NEPA categorical exclusion for the caisson experiment was granted by the<br/>Albuquerque Operations Office on 17 August. Efforts are underway to complete the<br/>lower-boundary condition apparatus and to fill the lower part of the caisson. We are<br/>waiting for the porous cups, which are on backorder from the manufacturer.<br/>Neutron probe tubes have been prepared for installation during filling of the caisson.Planned ActivitiesPrepare caisson for filling.<br/>Continue to fabricate lower-boundary condition apparatus as material becomes<br/>available.PublicationsNone

Preliminary data-do not reference

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Performance Assessment Calculational Support (WBS 1.2.1.4.7)

Activities and No activity reported this month. Accomplishments

#### WBS 1.2.3.1 Site Management and Integration

Site Management (WBS 1.2.3.1.1)

The objective of this task is to manage and integrate site characterization activities. Objective B. Carlos attended the SOC meeting at the Sample Management Facility. It was Activities and reported that SOC representatives visited the UZ-16 drilling operation. A potential Accomplishments conflict with striping of fractures was resolved during the meeting. E. Springer attended Integrated Test Evaluation (ITE) meeting in Las Vegas on 27 August to review the ITE report. Test Management and Integration (WBS 1.2.3.1.2) The objective of this task is to provide coordination for Los Alamos surface-based test Objective planning and package development. ESF Test Coordination. Initiated field construction and testing activities (as Activities and defined by TPP 92-07, "Fran Ridge Test Pit Mapping," and its associated job package, Accomplishments JP 92-7.) Record packages were submitted to the local records center. Surface-based Test Coordination. Continued to merge our administrative data base with YMP GIS to identify and track Los Alamos interfaces with other site characterization surface-based testing activities. Continue support of Los Alamos surface-based site characterization activities in **Planned Activities** response to Project program directives.

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

The purpose of this activity is to define the important mineralogical and geochemical Objective 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. Staff continued to write operating procedures and develop software for the INEL Activities and microdiffractometer. Accomplishments The revised paper, "Calcite deposits in fractures of drill cores USW G-2 and USW GU-3/G-3 at Yucca Mountain, Nevada," (milestone 3120) was submitted to the LANL TPO for policy review. B. Carlos and D. Vaniman visited the SMF from 31 August -1 September to examine the upper portion of drill core UZ-16 in order to select calcite and opal samples for further analysis during FY93. These samples will permit detailed sampling of the transition zone from micritic to sparry calcite. The report, "Geologic evaluation of six nonwelded tuff sites for a surface-based test

The report, "Geologic evaluation of six honweided unit sites for a surface-based test facility for the Yucca Mountain Project," (milestone 3137) is in revision. This report includes data on hydrologic properties (A. Flint, USGS) and mechanical properties (C. Rautman, SNL), as well as the mineralogical and chemical data collected at Los Alamos.

In fracture mineralogy studies, B. Carlos continued SEM studies of zeolites. Data currently being collected will be used to prepare a paper for the Zeolite '93 conference in Boise, Idaho, on 20–28 June. W. Cosgriff, a college student, has been hired to assist B. Carlos with analysis of fracture minerals.

## 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 None
- Milestone Progress 3120 30 June 1992 Calcite in the Upper Paintbrush Tuff Completed and submitted to TPO

3130 17 August 1992 Fracture Mineralogy of the Paintbrush Tuff

3137 30 September 1992 Geologic Evaluation of Six Nonwelded Tuff Sites for a Surface-based Test Facility for the Yucca Mountain Project In technical review.

#### Publications

B. Carlos, D. Bish, S. Chipera, and S. Craven Fracture-Lining Manganese Oxide Minerals in a Silicic Tuff Journal article, Chemical Geology Approved by YMPO; submitted for publication.

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 YMPO.

## WBS 1.2.3.2.1.1.2 Mineralogical 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 Staff transmitted responses by G. Valentine, B. Crowe, D. Vaniman, and S. Levy to the recommendations made in the National Academy of Science report, "Ground Water at Yucca Mountain: How High Can it Rise?" to the M&O. Published and in-progress work by D. Bish and G. WoldeGabriel was also cited.

Additional text and figures were supplied for the YMP topical report on calcitesilica deposits at Trench 14 and Busted Butte.

Electron microprobe analysis of altered rocks from Busted Butte and Yucca Mountain (Harper Valley) continued. Studies of these rocks will help resolve the question about recent hydrothermal activity around Yucca Mountain and will be incorporated in the final report on breccias and hydrothermal alteration.

Planned Activities Chemical and mineralogical characterization of samples from hydrothermal deposits exposed at the surface will continue.

We will contribute additional information to the YMP topical report on calcitesilica and breccia deposits. We will review the most recent unpublished report on groundwater discharge and alteration history by J. Szymanski.

Sample preparation and characterization of material for K/Ar studies will continue. G. Woldegabriel is preparing an invited presentation on K/Ar studies of zeolites for the Zeolite '93 conference.

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
65% complete. (Technical review complete.)

3142 31 July 1992 K/Ar Dating of Clays and Zeolites In technical review. 3143 30 April 1992 Experimental Dehydration of Volcanic Glasses In program review. 3150 15 April 1993 Final Report on Bedrock 30% complete. D. Bish and J. Aronson Publications Paleothermal and Paleohydrologic Conditions in Silicic Tuff from Yucca Mountain, Nevada Journal article, Clay and Clay Minerals Approved by YMPO. 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, Quaternary Research Approved by YMPO. D. Vaniman, D. Bish, and S. Chipera Dehydration and Rehydration of a Tuff Vitrophyre Journal article, Journal of Geophysical Research Submitted to YMPO. D. Vaniman, S. Chipera, and D. Bish Pedogenesis of Siliceous Calcretes at Yucca Mountain, Nevada Journal article, Science Submitted to YMPO.

## WBS 1.2.3.2.1.2 Stability of Minerals and Glasses

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 T Accomplishments

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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 A preliminary draft of Study Plan 8.3.1.8.1.2, "Physical Process of Magmatism and Effects on the Repository," has been forwarded to the DOE for submission to the NWTRB.

The first draft of the Issue Resolution Report was completed and will soon be submitted for internal review.

A video conference, the first of its kind between the DOE and NRC, was held with the NRC to discuss their comments and questions on Study Plan 8.3.1.8.1.1, "Probability of Magmatic Disruption of the Repository."

The age of the Q1<sub>3</sub> lava was obtained using the cosmogenic  ${}^{3}$ He method. The lava yielded duplicate ages of 64.6q7.4 and 72.8q8.6 ka, which is significantly older than an age of about 25 ka obtained using the thermoluminescence method.

Trace-element data for 40 from Lathrop Wells and Black Cone volcanic centers were analyzed by instrumental neutron activation analyses.

Chemical analyses of soils from trenches constructed on the north flanks of the Lathrop Wells volcanic center were obtained.

Detailed technical procedure (DP) 610, R0, "Extraction and Analysis of Noble Gases from Solids," was approved and distributed.

Work in Progress. Revised calculations were made using data from the newly recognized Pliocene basalt centers. Regression calculations of the latitude and longitude of vent sites for the Crater Flat volcanic zone were completed. Revised plots of cumulative magma volume versus time, which included the new data points, were completed.

Material was prepared for the September subcommittee meeting of the NWTRB.

The current status of U-Th disequilibrium measurements is as follows: a sample from Ql6 has been separated, dissolved, and the U-Th concentrations were measured; a duplicate sample from Ql4 has been separated and dissolved; a sample from Ql5 has been ground for mineral and phase separations; and a sample from Little Black Peak cone has been separated and dissolved.

Planned Activities Finalize the Issue Resolution report; it will be completed by 30 September.

A meeting with the NWTRB subpanel on tectonics is scheduled for 14-16 September in Las Vegas.

Trenching at the Lathrop Wells volcanic center is planned for the first and third weeks of September.

Problem Areas	We have delayed field mapping of the 3.7 Ma basalt centers of Crater Flat in order to concentrate on completion of the Issue Resolution Report.
	Because of time required to prepare for the NRC video conference and for the NWTRB meeting, the Issue Resolution Report will be delayed. It will be completed in September 1992.
Milestone Progress	3174 8 January 1992 Effects of Magmatic Disruption on the Repository (Study Plan 8.3.1.8.1.2, R0)
	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 First draft complete.
	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.

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## 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	Thirty-one samples were prepared by Hydro Geo Chem for chlorine-36 analysis by the University of Rochester and Lawrence Livermore National Laboratory. These samples were taken from the neutron-access bore holes, USW N37, N54 and N55, as well as Midway Valley soil.
	Hydro Geo Chem evaluated using an amperometric detector for improving the detection limit of the ion-chromatography system for bromide; unfortunately, the sensitivity and reproducibility of signals did not meet the needs of this activity. The effectiveness of concentrator columns will be determined next.
	Collection of cutting samples for $^{36}$ Cl analysis from UZ-16 and new neutron access bore holes continued.
Planned Activities	Staff will address study plan review comments; revise existing DPs; prepare new DPs; process soil samples for Cl/Br and chlorine-36/Cl ratios; process cuttings samples from neutron-access bore holes; and collect additional soil samples from Yucca Mountain area, as opportunities arise.
Problem Areas	None
Milestone Progress	3191 Procedure for Chlorine-36 Analysis of Unsaturated Zone Samples 30 September 1992 95% complete
Publications	None

Accomplishments

## WBS 1.2.3.3.1.2.5 Diffusion Tests in the ESF

- ObjectiveThe objective of this task is to determine *in situ* the extent to which the nonsorbing<br/>tracers diffuse into the water-filled pores of the Topopah Spring welded unit.Activities andNo significant activity in this study.
- Milestone Progress No Level II milestones are planned for 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-25c #1, UE-25c #2, and UE-25c #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 Software Qualification. Z. Dash and B. Robinson continued to serve as temporary Software Configuration Manager and CCB Chair, respectively.

Detection of Microsphere Concentrations. B. Newman completed a preliminary study on the feasibility of using a Los Alamos flow cytometry facility to measure the concentration of fluorescent microspheres. Results appear to be very promising and suggest that this method should be developed further. The instrument, ordinarily used in biological research to sort and isolate individual cells, is very selective, it being able to detect small microsphere concentrations in the presence of very high background colloid concentrations (much higher than would be present in laboratory or field samples). A letter report has been written describing this work, with recommendations for ways to improve the technique.

Modeling. No significant progress this month because we have been required to spend a large quantity of time on the SQA effort.

A draft of B. Robinson's paper to be submitted to the *Journal of Radioactive Waste Management* (special issue on the Yucca Mountain Site Characterization Project) has been completed and is in internal technical review.

Planned Activities Continue to contribute to the SQA effort by serving as temporary Software Configuration Manager (Z. Dash) and temporary CCB Chair (B. Robinson).

Complete documentation of batch-sorption experiments with lithium bromide.

- Continue modeling studies using FEHMN to support the design of the field tests.
- Problem Areas We were unable to make significant progress on our modeling efforts this month because of the large amount of time we devoted to the SQA effort.

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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 personnel reassignment. 3196
	27 July 1992 FRACNET Documentation Delayed due to change of assignment for principal investigator.
	R529 Evaluation of Preliminary Application of FEHMN to Yucca Mountain Completed.
Publications	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.
	W. L. Polzer and 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 Accepted.

#### 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 Study Plan. Review of comments and revision of the Groundwater Chemistry Model Study Plan, R0, continued.

Other Activities. The matrix for investigating most-active groundwaters was refined. The objective is to categorize groundwaters by compositional variables that affect radionuclide solubility and/or sorption properties. Variables of interest presently are measured Eh potential, pH, and bicarbonate content. EQ3/6 is being used to conduct simulations using the different kinds of groundwater compositions. A letter report describing the results of this exercise is due 30 September and is on schedule.

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 study plan.

Complete letter report on most-active groundwater.

Continue USGS collaboration. Delays in material and equipment for the USGS downhole 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

Milestone Progress 3006 31 August 1992 Eh and pH Buffering Capacity 3415 30 September 1992 Most-Active Groundwater Chemistry

Publications

None

## 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	Experiments to measure the batch-sorption coefficients of neptunium on crushed-tuff samples under a variety of pH, pCO <sub>2</sub> , and water compositions were started by KH. Kung with the assistance of C. Overly. The rock samples and Np-traced solutions will be allowed to react until 15–17 September.
	Experiments were started to evaluate the properties of a new type of filter for separating the solution and solid (+colloidal) phases after reaction in a batch-sorption determination. The filters used previously for this purpose are no longer available.
	BET surface area results for -40 samples have been obtained from Quantachrom Corp. for use in the study of sorption coefficient dependence on particle grinding. Results show that there is little dependence of surface area on the rock particle size down to 38 mm diameter, indicating that both the zeolitized- and devitrified-tuff samples studied have "accessible" surface area that is controlled by features (pores, exchange sites, open grain boundaries, microfractures) of a spatial scale smaller than this.
	M. Hawley has obtained the first atomic resolution images of geothite, an iron oxy- hydroxide mineral with extremely high surface sorption capacity for many cations. Geothite is expected to be an important sorbing component of fracture linings in Yucca Mountain. Its sorption behavior can also be compared with that of hematite to provide important insights into possible sorption mechanisms. To our knowledge, no one has yet published such high resolution AFM scans of geothite.
Planned Activities	Batch-sorption experiments for neptunium will be permitted to react until mid- September, when the solution and solids will be separated and prepared for scintillation counting.
	Tests of the new type of filter will be completed.
	We will prepare the milestone report covering the dependence of sorption coefficients on particle size and water composition, now that important surface area measurements have been made.
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

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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-12325-C, 1992) Published.

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## 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	As reported previously, staff has been investigating the dissolution of hematite by <i>Pseudomonas</i> sp. Results to date have indicated that the presence of microorganisms does increase mineral dissolution.
	Unsaturated flow columns are now being constructed to study microbial activities in unsaturated environments, specifically establishing long-term sterile conditions within a soil column.
	Work continued on milestone 3080, "Chelation," and 3092, "Colloidal Agglomeration."
Planned Activities	Continue plutonium K <sub>d</sub> experiments.
	Continue colloidal agglomeration experiments.
Problem Areas	None
Milestone Progress	3080 30 September 1992 <i>Report on Chelation</i> In preparation.
	3092 30 September 1992 <i>Report on Colloidal Agglomeration</i> In preparation.
	3176 30 September 1992 Procedure for Determination of Formation Constants In progress.
	3177 30 September 1992 <i>Procedure for Determination of Effects on Colloidal Agglomeration</i> 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 We completed the study plan revision, and all comments have been addressed. We also completed the Validation of Estimates phase of the planning and scheduling activity, identifying 16 discrete study plan activities.

Speciation Studies. Results of our plutonium carbonate NMR studies are being written up and will be submitted to the *Journal of the American Chemical Society*.

We prepared Np(V) solutions for sorption studies; processed Pu(III) NMR data; and reprocessed Pu residues for new experiments.

D. Clark and P. Palmer have developed a technique for growing single crystals of Pu(IV) carbonate complexes for study by x-ray crystallography and solution NMR. This first-time structural analysis of Pu(IV) carbonate complexes will provide valuable speciation information about them.

We plan to conduct several additional PAS experiments to address the issue of the number of species present in the solutions. We have collected a small quantity of additional data in a new wavelength range and will look for traces of specific oxidation states in this range. These experiments will be completed in approximately one month.

We continued to write Milestone Report 3031, "Plutonium(IV) and Plutonium(VI) Carbonate Speciation Studies by NMR and PAS Spectroscopies"; it will completed by 30 September.

Solubility Studies. We continued determining the solubility of Np, Pu, and Am in UE-25p #1 water at 60°C. The Np undersaturation experiments at pH 7 again showed pH-dependent concentration variations as reported in June 1992. (Because we measured a higher Np concentration at pH 6.78 than at pH 7.00, we have noted the measured pH along with Pu(IV) concentrations in Table II.) Because the three Np undersaturation experiments yielded the same results as in the oversaturation experiments, they will be stopped in September, which will allow them to run for the same length of time as the oversaturation experiments.

Two samples from each of the neptunium oversaturation experiments, a dried unwashed sample and a sample washed with small amounts of water and ethanol before drying, were prepared for x-ray powder diffraction measurements. The powder patterns were compared with the following published patterns: a) orthorhombic Na<sub>0.6</sub>NpO<sub>2</sub>(CO<sub>3</sub>)<sub>0.8</sub>•2H<sub>2</sub>O, b) orthorhombic Na<sub>0.6</sub>NpO<sub>2</sub>(CO<sub>3</sub>)<sub>0.8</sub>•nH<sub>2</sub>O, and c) monoclinic Na<sub>3</sub>NpO<sub>2</sub>(CO<sub>3</sub>)<sub>2</sub>•nH<sub>2</sub>O. The washed and unwashed solids from the pH 6 experiment matched orthorhombic Na<sub>0.6</sub>NpO<sub>2</sub>(CO<sub>3</sub>)<sub>0.8</sub>•2H<sub>2</sub>O very well. The unwashed solids from the pH 7 experiment matched orthorhombic Na<sub>0.6</sub>NpO<sub>2</sub>(CO<sub>3</sub>)<sub>0.8</sub>•nH<sub>2</sub>O very well, and the washed solid matched orthorhombic Na<sub>0.6</sub>NpO<sub>2</sub>(CO<sub>3</sub>)<sub>0.8</sub>•2H<sub>2</sub>O well. At pH 8.5, the washed and unwashed solids matched orthorhombic Na<sub>0.6</sub>NpO<sub>2</sub>(CO<sub>3</sub>)<sub>0.8</sub>•2H<sub>2</sub>O very well. Each of the powder

#### Activities and Accomplishments (continued)

patterns from the experiments at 60°C, both washed and unwashed, was also compared to the powder pattern of the solid produced in the experiment using UE-25p #1 water at 25°C and pH 6; all of them fit this pattern extremely well. Oxidation state determinations were performed on the two remaining plutonium oversaturation experiments at pH 7 and 8.5. The results are listed in Table I, along with those obtained last month for the pH 6 experiment.

#### Table I. Plutonium Oxidation State Distribution at Steady-State in UE-25p #1 Water at 60°C at pH 6, 7, and 8.5

Final Oxidation State Distribution	Initial Pu(IV) Concentration at pH 6, Day 258 (in %)	Initial Pu(IV) Concentration at pH 7, Day 273 (in %)	Initial Pu(IV) Concentration at pH 8.5, Day 278 (in %)
Pu(IV) Polymer Pu(III) Pu(IV) Pu(V) Pu(V)	$(0.4 \pm 0.1) (0) (1.3 \pm 0.4) (4.2 \pm 0.6) (94.1 \pm 11.1)$	$(1.8 \pm 0.5)$ (0) (0.6 ± 0.5) (5.1 ± 1.1) (92.5 ± 11.2)	$(1.0 \pm 0.1) (3.7 \pm 2.0) (9.6 \pm 1.2) (0) (86.1 \pm 11.7)$
Total Pu Conc. ( <u>M)</u>	$(9.42 \pm 0.84) \ge 10^{-8}$	(1.07 ± 0.10) x 10 <sup>-7</sup>	$(7.00 \pm 0.63) \times 10^{-7}$

The results indicate Pu(VI) as the predominant species formed in UE-25p #1 water at 60°C, whereas at 25°C the predominant species was Pu(V). In J-13 water at 25°C, the predominant species was also Pu(V) (63–73%), but at 60°C the percentage of Pu(VI) rose significantly, especially at pH 6 and 7 (72 and 52 % respectively). The oxidation state changed from predominantly Pu(V) to Pu(VI) when the temperature is increased from 25°C to 60°C in both waters. The higher carbonate content in UE-25p #1 water may provide stronger carbonate complexation in solution, stabilizing the Pu(VI) in UE-25p #1 water at the same temperature.

After obtaining oxidation state determinations, the three plutonium oversaturation experiments were stopped, and small portions of the solids were removed for x-ray diffraction studies. The remaining solids were used to start Pu undersaturation experiments the same way the americium/neodymium and Np undersaturation experiments were started (as explained in the letter report for March 1992, TWS-LBL-03-92-02). Each of the three experiments was sampled on day zero and day one and will continue to be sampled until steady-state is reached and maintained for some time. On day one, the aqueous plutonium concentration in all three experiments ranged from  $3 \times 10^{-8}$  to  $6 \times 10^{-8}$  M, already very near the steady-state values obtained in the oversaturation experiments.

The most recent sample of the americium/neodymium experiment at pH 7 yielded a concentration almost an order of magnitude higher than the previous sample. This concentration has varied randomly over an order of magnitude between samplings, and its cause is under investigation. (This experiment also shows sampling-related problems for determining aqueous americium/neodymium concentrations.)

Activities and Accomplishments (cont.)	The results from oxidation state determinations for the pH 8.5 americium /neodymium experiment are being re-examined. Lanthanum fluoride co- precipitation results showed americium in a higher oxidation state such as V or VI; however, the yield of americium in the separation was significantly higher than 100%. Therefore, either some higher oxidation state americium exists in the pH 8.5 experiment, or the americium activity in the one fraction came from an external contamination after the co-precipitation separation. The oxidation state determinations are being repeated to resolve this discrepancy.
	Regarding the LBL QA effort, the focus was on preparations for the yearly Los Alamos YMP audit. Three classroom training sessions, including written tests, were held.
	The draft detailed technical procedure (DP) for balance calibration was updated to comply with the new QP for Measuring and Test Equipment. Review comments are currently being addressed for other three DPs, "Concentration Determination of Soluble Radionuclides from Data Provided by a Low-energy Gamma-ray Counting System" (LANL-LBL-DP-01), "Calibration of Low-energy Gamma-ray Counters," and "X-ray Powder Diffraction Analysis." They will be sent out for review in the near future.
Planned Activities	Efforts in all areas will continue.
Problem Areas	Alpha radiolysis effects have hampered our analysis of both 13-C and 17-O NMR data. We have devised a new sample tube, which requires a reversal of our double- containment apparatus for work with intensely alpha-active actinides; we plan to test this new apparatus in the next few months.
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 <i>Plutonium(IV) and Plutonium(VI) Carbonate Speciation Studies by NMR and PAS</i> <i>Spectroscopies</i> On schedule.
	3329 30 September 1992 Report on Neptunium, Plutonium, and Americium Solubility Experiments in UE-25p #1 Water from Oversaturation. On schedule.
	3330 1 January 1993 <i>Evaluation of Alternative Detection Schemes in Photoacoustic Spectroscopy</i> Early completion anticipated.
	Letter Report Spectroscopic Studies of the Hydrolysis of $UCl_4$ : Spectral Effects of Ligand Exchange In preparation.

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Preliminary data-do not reference

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 Co.nplexes 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.
	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. E. Morris, C.D. Tait, S. A. Ekberg, and P. D. Palmer Speciation of Plutonium in Carbonate Media Conference abstract, Materials Research Society Approved by YMPO.
	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 <i>Measured Solubilities and Speciations of Neptunium, Plutonium, and Americium in a</i> <i>Typical Groundwater (J-13) from the Yucca Mountain Region</i> LA-series report Submitted to YMPO.
	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, S. A. Ekberg, and P. D. Palmer, and D. E. Morris Plutonium Carbonate Speciation Changes Journal article, Inorganic Chemistry In preparation.

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# 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 Np transport work using crushed-tuff columns made from tuffs G4-1530.3 and G4-275. We began eluting Np in J-13 water through two columns made with tuff G4-1530.3. The flow characteristics of these columns were reported last month.

We continued Np batch-sorption experiments with tuffs G4-1530 and G4-275 in USWH-3, UE-25p #1, and J-13 waters to support our transport experiments. Specifically, we conducted batch-sorption experiments with Np solutions and pure mineral separates in hematite, montmorillonite, clinoptilolite, and quartz. We also completed equilibration of these tuffs and mineral separates and began adding Np to the pre-equilibrated tuff and minerals.

Staff continued organizing the colloid workshop to be held in early 1993.

Four detailed technical procedures were implemented.

Staff submitted five technical data information forms summarizing dynamic transport and diffusion studies to the Automatic Technical Data Tracking System.

- Planned Activities Continue all work discussed 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-12325-C, 1992) Published.

### 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 Certification of TRACRN continued. The Design Phase Baseline was resubmitted in response to RIDS generated by the TRACRN review committee. Verification continued and is nearly complete. Staff found that TRACRN agreed well with the analytic and other numerical solutions being used for the verification exercise. These comparisons were being documented in the Verification and Validation Report. The user's manual is nearly complete.

Certification of FEHMN and GZSOLVE continued. G. Zyvoloski worked with B. Robinson and Z. Dash (C-Wells Reactive Tracer Task) on their documentation and verification.

- Planned Activities Continue certification of TRACRN.
- Problem Areas None
- Milestone Progress 3052 30 September 1992 Baseline Documentation for TRACRN

PublicationsK. Birdsell, K. Eggert, and B. Travis<br/>Three-Dimensional Simulations of Radionuclide Transport at Yucca Mountain<br/>Journal article, Radioactive Waste Management and the Nuclear Fuel Cycle - Special<br/>issue on the Yucca Mountain Project<br/>Approved by YMPO; submitted.

K. Birdsell, K. Campbell, K. Eggert, and B. Travis Sensitivity Analysis of Integrated Radionuclide Transport Based on a Threedimensional 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-12325-C, 1992) Published. \_\_\_\_\_

## 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 analogs will be assessed for their potential to provide the required data.
Activities and Accomplishments	E. Springer attended the Exploratory Studies Facility's (ESF) test co-ordination meeting in Las Vegas on 18 August. Issues discussed included FY93 plans for the ESF and experiences with implementing field tests at other sites (Basalt and WIPP).
	A presentation on proposed Los Alamos field-testing activities for radionuclide transport was made to B. Delakowik from Germany on 24 August in Las Vegas.
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 In press.

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#### 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 Los Alamos continued to support closure reports for both the calcite-silica and erosion issues.

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. We are awaiting a response from YMPO.

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 and submitted to NRC for review.

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 was completed in July 1992.

Groundwater Chemistry Modeling, R0 (8.3.1.3.1.1). In March 1991 this study plan was submitted to the project office for review. Comments were returned 31 May 1992 and are now being addressed.

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. YMPO returned study plan in August 1992 for final word processing revisions.

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 Analog 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. The review was completed in August and returned to the principal investigator for comment resolution.

**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 were submitted in August 1991.

Study Plans (continued) Dissolved Species Concentration Limits, and Colloid Formation and Stability, R0 (8.3.1.3.5.1; 8.3.1.3.5.2). In November 1991, 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 September 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 September 1992.

Retardation Sensitivity Analysis, R0 (8.3.1.3.7.1). This study was approved by the DOE and sent to the NRC for review in July 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. The internal review was completed in August and sent to the principal investigator for comment resolution.

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 Continued gathering information on Tracers, Fluids, and Materials (TFM) and requested waste isolation impact and test interference analysis for TFMs from CRWMS M&O and SNL.

Continued to support M&O efforts to develop a position paper on prototyping.

Prepared briefings for biweekly ESF management meetings, attended biweekly ESF Engineering Development Division (ED&D) meetings, and participated in ESF excavation and ESF testing planning meeting.

Reviewed Title II design, Package 1A.

## Planned Activities Continue to develop definitive design related information for launch-chamber tests.

Continue to support integration meetings such as ESF design, surface-based testing and its interface with ESF testing, TIG, and SMF.

Support ED&D justification for prototype test facility and ESF budget options and strategies. Develop interfaces for testing and the ESF design.

Revise and update the PSAR as required.

Begin identifying Project Integrated Data System (IDS) planning.

Prepare abstract for the upcoming waste management conference.

Finalize abstracts for papers for High-Level Waste Management Conference, 34th Rock Mechanics Symposium in Madison, Wisconsin.

Prepare paper on the Integrated Data System for ESF to be submitted to the for the Second International Symposium on Mine Mechanization and Automation.

Problem Are	eas Non	e
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Milestone Progress No milestones for FY91.

Publications None

### WBS 1.2.6.8.4 Integrated Data System

This activity has been deferred.

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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

Preliminary data-do not reference

#### 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 Eighty records and/or records packages were received by the RPC; seven of these were returned to the originators for corrections.

The CRF rejected five records and/or records packages. The RPC resubmitted four previously rejected records to the CRF.

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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	Software. Two Configuration Control Board meetings were held. A status report was distributed, and committees for software reviews were established.
	Grading Reports. All Los Alamos grading reports have been approved by the Project Office.
	<b>Records/Document Control.</b> Quality Administrative Procedure (QP) QP-02.9, R1, "Personnel Proficiency Evaluations"; QP-02.11, R1, "Personnel Orientation and Detailed Technical Procedures"; detailed technical procedure (DP) DP-15, R3, "Crushed Rock Column Studies"; DP-60, R3, "Preparation of NTS Samples for LANL YMP Solid Core Experiments"; DP-61, R3, "Solid Rock Column Experiment," DP-63, R3, "Preparation of NTS Core Samples for Crushed Rock Experiments"; and DP-610, R0, "Extraction and Analysis of Noble Gases from Solids," were approved and distributed. DPs 26, 27, 64, 74 and 82 were withdrawn.
	Training. We are currently evaluating the LaDelfe training data base program. The new orientation program, which was scheduled for presentation at Lawrence Berkeley Laboratory in August, has been postponed because of scheduling conflicts.
	<b>Program Development.</b> Twenty QPs were in various stages of revision. The 2001 quality assurance budget was revised according to DOE guidelines; basis of estimates were completed. The YMPO draft Qualified Suppliers List was reviewed.
	Audits/Surveys. Responses to corrective action reports CAR-057 and CAR-058 were submitted to YMPO; YMPO staff have accepted the proposed resolution for corrective action report CAR-057. The LANL-AR-92-09 (Stanford University) and LANL-AR-92-10 (Lawrence Berkeley Laboratory) audits were postponed because of scheduling conflicts. The annual management assessment report of the Los Alamos QA program is in TPO review.
Planned Activities	Internal audit and survey reports will be completed. QP revisions will continue. A draft organizational procedure will be written and submitted for informal review, and the notebook procedure (QP-03.5) will be submitted for formal review.
	The selection of a new software configuration manager will be delayed until budget uncertainties have been resolved. The audit schedule will be revised to accommodate scheduling conflicts.
Problem Areas	The selection of a new software configuration manager will be delayed until budget uncertainties have been resolved. The audit schedule will be revised to accommodate scheduling conflicts.
Milestone Progress	No milestones for FY91.

#### Publications

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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, Proceedings of the ASQC Energy Division Annual Meeting Approved by YMPO.

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