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PNL-5015-2

(FY-84)

Uranium Recovery Research Sponsored by the Nuclear Regulatory Commission at Pacific Northwest Laboratory

Quarterly Progress Report January to March 1984

May 1984

Prepared for the U.S. Nuclear Regulatory Commission
under Contract DE-AC06-76RLO 1830
NRC FIN B2269, B2292, B2370, B2379

Pacific Northwest Laboratory
Operated for the U.S. Department of Energy
by Battelle Memorial Institute

Battelle

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PACIFIC NORTHWEST LABORATORY
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URANIUM RECOVERY RESEARCH SPONSORED BY THE NUCLEAR REGULATORY COMMISSION AT PACIFIC NORTHWEST LABORATORY

QUARTERLY PROGRESS REPORT
JANUARY TO MARCH 1984

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Pacific Northwest Laboratory
Richland, Washington 99352

EXECUTIVE SUMMARY

This report documents progress for four major research projects which include 9 discrete research tasks and a management task being conducted for the U.S. Nuclear Regulatory Commission (NRC), Office of Research, Waste Management Branch. The primary purpose of these tasks is to provide information to help the NRC license uranium recovery facilities. A truncated title of each task and a brief highlight summary are provided here.

- Long-Term Stabilization

Hydrologic modeling of the Dolores River at the Slickrock site was completed during the quarter. The modeling included the computation of hydraulic design data for five flood events. Work continued on the analysis of rock riprap design methods for the Slickrock site.

The reports for subtasks E (Economics of Riprap Design Procedures) and H (Impoundment Slope Stability and Settlement) were completed in draft form. The Subtask E report was reviewed, revised and forwarded to clearance prior to preparation of a camera-ready copy. The report for Subtask H is undergoing revision. The report for Subtask D (Vegetation Covers) was published.

Three technical papers were presented at the Sixth Symposium, Management of Uranium Mill Tailings, Low-Level Waste and Hazardous Waste, held at Ft. Collins, CO, February 1-3, 1984. The papers presented results from the vegetation cover study and the hydrologic analysis of riprap design methods.

- Interim Stabilization of Mill Tailings Piles

A paper on the field test work was presented in February at the Sixth Annual Symposium on Uranium Mill Tailings Management, Low-Level Waste, and Hazardous Waste held at Colorado State University. A review article on fugitive dust control was submitted to the journal Nuclear Safety for publication. A report on the laboratory studies was completed and sent to NRC for printing. Data from the PNL windscreen field test is being collected and analyzed to determine the effectiveness of windscreens in reducing fugitive dust.

- Tailings Dewatering Techniques

Measurements of the required saturated material properties (i.e., void ratio and hydraulic conductivity) as functions of the effective stress have been successfully completed for all three materials from the Grand Junction mill tailings pile (sand, slime, and a sand/slime mix). The partially saturated measurements are currently in progress. The lab apparatus has been further modified to improve the measurements. Due to the length of time necessary to run the partially saturated experiments, the major effort is being concentrated on the material expected to consolidate the most from dewatering: the slimes.

The two stress-state description of void ratio, saturation, and conductivity in the partially saturated zone was tested. It became evident that retaining classical consolidation theory in the saturated zone was inappropriate, since our void ratio versus effective stress data did not fit a log-linear relation. The model was then changed so that consolidation in the saturated zone could be calculated from the void ratio versus effective stress relationship derived from the laboratory data.

Testing of partially saturated consolidation will continue using the Grand Junction slime data that have been generated to date, supplemented by data from the literature.

- Tailings Neutralization and Other Alternatives in Immobilizing Toxic Materials in Tailings

This task began with a review of the existing literature on neutralization techniques. The results of the review indicated that no single technique is 100% effective in the treatment of uranium mill tailings solution. Neutralization of the solution fraction of the tailings is the most inexpensive method of treatment reviewed. The high cost of solidification techniques may make the treatment of bulk tailings impractical. Specific ion removal processes may be necessary for those elements that are highly soluble or have a low allowable maximum concentration limit.

- Evaluation of Seepage and Leachate Transport From Tailings Disposal Facilities

The evaluation method developed under this project and document as NUREG/CR-3560 during the previous quarter has been used to better understand the range of conditions that may occur when uranium tailings are disposed in deep underground mine stopes. The environmental consequences of tailings disposal in abandon mine tunnels are being analyzed by considering tailings leachate transport by regional ground water away from and into downgradient water supply wells. Specifically, the concentration of leachate constituents in the water pumped from the well is the measure being used when sufficient water can be realistically obtained from a well in the regional aquifer. Considerable progress has been made in understanding the interaction of the flow system parameter upon configuration of the uranium tailings leachate plume downgradient from the mine stope.

- Effluent and Environmental Monitoring Methods and Equipment and Instrument Testing

This task was not funded in FY-84, and exhausted its funds carried-over from FY-83 in December. No work was performed in the last quarter except the completion of topical reports on well-logging techniques and comparison of radon fluxes and preparation of letter status reports on work completed previously in several other subtasks.

- Attenuation of Radon Emissions

Camera ready copies of an engineering handbook to calculate radon attenuation for uranium mill tailings cover design and a document in the format of an NRC Regulatory Guide were delivered to NRC. Another report, testing the validity of the Handbook methods on data from the Grand Junction tailings pile, will be delivered next quarter.

- Assessment of Leachate Movement From Uranium Mill Tailings

Two types of experiments are currently being conducted. In the first type, leaching experiments were designed with various column lengths and diameters. Synthetic ground water was passed through these columns after packing them with uranium mill tailings. Results indicate that there are no effects on the effluent concentrations of the leaching columns due to differences in column lengths (residence time) or column volumes (scaling). The results of these experiments provide a foundation for using column experiments to predict long- and short-term movement of contaminants from tailings impoundments because of leaching due to migrating ground waters or rainfall.

The objective of the second study is to evaluate the effects of residence time when acidic uranium mill tailings solutions contact coarse sediments. The column lengths varied from 1:1 to 8:1. The varying lengths of columns allowed different solution/sediment contact times. All conclusions drawn thus far with regard to these columns are very tentative. More definitive interpretations will be possible once all data are obtained.

Speciation/solubility modeling has been performed on the solutions obtained from 17 monitoring wells at an evaporation pond in Wyoming. The conceptual chemical model of the field site has been formulated. This conceptual chemical model will be used along with the MINTEQ computer code to predict contaminant concentrations with distance.

- Methods of Minimizing Ground-Water Contamination in In Situ Leach Uranium Mining

The natural restorative capacity of an *in situ* uranium leach mine in Wyoming was studied in the laboratory using aquifer sediments and synthetic lixiviant. Contrary to the results of a similar experiment using solutions and sediments from a Texas mine site, we found that for the Wyoming case uranium movement through the sediment was not significantly retarded by water/sediment interactions. Analyses of pyrite in the sediment showed that the Wyoming sediment used in the experiment had much less pyrite than the Texas sediment, and, because pyrite may affect the reducing capacity of the sediment and thereby limit the concentration of uranium in solution, its variation in the sediments may be the important factor producing differences in the experiments. Additional work needs to be done to more fully evaluate the natural restorative capacity of the Wyoming sediments.

The effect of ground-water sweeping on aquifer restoration was studied in the laboratory with sediment, lixiviant, and ground water from a Texas leach mine. Solution and sediment chemistry data show that residual uranium minerals in the leach zone may be dissolved during sweeping. This will maintain elevated uranium concentration in solution which will inhibit restoration of the aquifer. The potential contaminants arsenic, selenium, and molybdenum will be at least partially immobilized by the sediments during sweeping and this will enhance restoration for these elements.

CONTENTS

INTRODUCTION

Pacific Northwest Laboratory (PNL) is currently conducting research for the U.S. Nuclear Regulatory Commission (NRC) on uranium recovery process wastes for both active and inactive operations. The overall objective of this research is to provide NRC and their licensees with technical guidance on several issues related to management of wastes from uranium mills and in situ recovery operations. Principal issues addressed in these studies are: designs and performance of radon-suppression covers; the incentives and constraints for using protective covers, and designs for armoring tailings pile radon-suppression covers; short-term stabilization options for controlling windblown particles; leachate movement in soil; tailings dewatering and consolidation; disposal deliberately below the water table; neutralization incentives; contamination control and restoration in in situ uranium recovery; and environmental measurements, instrumentation, and protocols for assessing the radioactive contamination associated with uranium milling and mill tailings. Results of these studies will provide means to better evaluate the environmental impacts during and following the active life of a uranium recovery facility, and many will be used in developing regulatory guides.

The National Research Council Committee on Onshore Energy Minerals Management Research (COEMMR) stated:

> "The most significant unresolved (uranium) milling-related problem appears to be the safe disposal of radioactive and heavy metal-laden mill wastes. Potentially suitable tailings management techniques, using different approaches, have been adopted at several of the mills commissioned in the last few years. It is important that field performance of both old and new techniques be monitored and the data critically examined to determine the effectiveness of the different methods, and the nature of any required modifications."

NRC-sponsored uranium recovery research at PNL is focused on NRC regulatory responsibilities for uranium-recovery operations:

- license active milling and in situ extraction operations,
- concur on the acceptability of DOE remedial-action plans for inactive sites,
- license DOE to main inactive sites following remedial actions

PNL's program consists of four coordinated projects comprised of a program management task and nine research tasks that address the critical technical and safety issues for uranium recovery. Specifically, the projects endeavor

(a) Information available in Proceedings of the Workshop on Research and Information Needs for Management of Uranium Production from Leased Federal and Indian Land, held December 9-10, 1982, Albuquerque, New Mexico. In publication through the Committee on Onshore Energy Mineral

Management Research sponsored by the National Research Council, Washington, D. C.

to find and evaluate methods to:

- prevent erosion of tailings piles and prevent radon release from tailings piles
- evaluate the effectiveness of interim stabilization techniques to prevent wind erosion and transport of dry tailings from active piles
- estimate the dewatering and consolidation behavior of slurried tailings to promote early cover placement
- design a cover-protection system to prevent erosion of the cover by expected environmental stresses
- reduce seepage into ground water and prevent ground-water degradation
- control solution movement and reaction with groundwater in in situ extraction operations
- evaluate natural and induced restoration of groundwater in in situ extraction operations
- monitor releases to the environment from uranium recovery facilities.

This quarterly progress report for the uranium recovery research program at PNL covers the research performed for the NRC at PNL in the period January, 1984 through March, 1984. The following sections report the progress in uranium recovery research at PNL during the quarter.

This information is preliminary, and conclusions should be regarded as tentative. Further information will be contained in the topical reports published, or scheduled to be published throughout this research effort. The authors solicit comments and suggestions from readers of this progress report.

STABILIZATION, ENGINEERING, AND MONITORING ALTERNATIVES ASSESSMENT FOR IMPROVING REGULATION OF URANIUM RECOVERY OPERATIONS AND WASTE MANAGEMENT

Project Manager: M. G. Foley

FIN B-2370

OBJECTIVE

This research effort will address many of the technical issues related to providing and assessing methods for predicting, evaluating, and limiting the environmental effects of wastes from uranium milling operations. The principal objective of the project is to provide information essential in licensing actions to ensure that individuals and the public will be adequately protected during the active life of the mill and over an indefinite time period following shutdown. The scope of this investigation embraces seven tasks, each of which will be discussed separately in the report.

TASK 1. LONG-TERM STABILIZATION

Task Project Manager: W. H. Walters
Principal Investigator: W. H. Walters
M. G. Foley
R. L. Skaggs
R. M. Ecker
P. A. Beedlow
C. S. Kimball

FIN B-2370 (Task 1)

OBJECTIVE

Through this task we will provide the Nuclear Regulatory Commission with an engineering handbook on the protection of decommissioned uranium tailings impoundments from long-term erosion by wind and water. The handbook will also include a technical/economic evaluation of riprap design procedures, an evaluation of watershed models for estimating potential surficial erosion around the tailings pile, and criteria to estimate embankment slope stability.

ACCOMPLISHMENTS DURING PAST QUARTER

Subtask A. Technical Feasibility Study

Work continued on the hydraulic analysis of the Dolores River at the Slickrock site. Hydraulic design data were computed for five potential flood events including the estimated probable maximum flood (PMF) and the 100 yr. and 500 yr. flood discharges. Since a computed standard project flood (SPF) discharge was not available, the PMF was increased by 25 and 50 percent to provide two additional discharges for comparison. The flood events and discharges are as follows:

Flood Event	Discharge (cfs)
100 Yr	14,000
500 Yr	18,000
PMF	46,100
PMFx1.25	57,625
PMFx1.50	69,150

Since there were no observed water surface profiles available from previous floods, the Mannings n roughness values were varied for the both the in-channel and overbank PMF discharges. The purpose of varying the Mannings n was to test the sensitivity of the hydraulic design data on these values. Because the Mannings n values are selected based on observed river and flood-plain characteristics the parameter is subject to individual interpretation and, therefore, can easily be disputed. Also, as a general rule the roughness values decrease as the flood discharge increases.

Theselected n-values for the Slickrock site were both increased and decreased by 10, 25, and 50 percent and hydraulic data computed for the PMF event (46,100 cfs).

Subtask B. Riprap Design for Overland Erosion and Local Scour

No work was performed on this subtask during the quarter. The literature review has been completed. Originally, the results of this subtask and those of subtask C were to be included in the subtask A report. Current plans are to publish both the subtask B and C results together in a separate report to provide more visibility for the work.

Subtask C. Riprap Protection for Local Drainage Channels

No work was performed on this subtask during the quarter. The literature review has been completed. The results will be published in a report with those of subtask B as discussed above.

Subtask D. Vegetation Covers

The final report for this subtask was completed and published during the quarter.

Subtask E. Economics of Riprap Design Procedures

The report for this subtask was completed in draft form for review during the quarter. Preliminary results are as follows:

The study results indicate that riprap design parameters such as side slope angle and specific gravity of the rock can have optimum values or ranges for a specific tailings site. An added safety factor to the median rock size can significantly increase costs but the increase is less site specific.

By using the Safety Factor Method for riprap design, the optimum embankment side slope in terms of costs can easily be determined for the design flood conditions. For the Grand Junction and Slickrock case studies the optimum side slope is about 5H:1V. Above this value the rock size requirement increases significantly.

The cost evaluation indicated that the use of local rock, even though it has a lower specific gravity, could be more economical than using denser rock located farther from the site. For the Grand Junction site a cost savings of up to 45 percent could be realized by using local Dakota sandstone (specific gravity = 2.25) instead of the nearest limestones (specific gravity = 2.70) which are about 80 miles from the site.

Subtask F. Watershed Model Evaluation

Work on this subtask commenced during the quarter.

Subtask G. Rock Selection and Durability

The final product of this subtask, to be delivered to NRC in December, 1984 as part of the final stabilization handbook, will be a procedure for selecting and testing rock for armoring uranium tailings piles, and recommendations for techniques to account for long-term chemical weathering and for over-sizing less-durable rocks. Laboratory work for this subtask was completed during the last quarter, and preparation of a topical report partially completed. Results suggest that a two-part selection and testing procedure be used, as follows:

Reconnaissance Phase - Identify potential sources and rank them to focus on the most desirable.

Use published topographic and geologic maps and stratigraphic descriptions of site area to identify resistates and select potential candidates.

Use aerial photographs in conjunction with maps to select potential sampling sites, emphasizing the potential for geomorphic features that provide for determination of relative and semi-quantitative absolute weathering durations for samples.

Collect samples, using published field techniques to rank apparent durability at the outcrop and cull any obviously unsuitable lithologies.

Test the samples in the laboratory using the slake abrasion and wetting/drying tests to rank their resistance to environmental stresses and to determine their weight loss as a function of number of test cycles. The slake abrasion is analogous to flood abrasion, and can be used as a measure of rock durability for rip rap.

This phase of testing ranks the samples on the basis of their mechanical durability and provides an estimate of their expected weight loss as a function of number of cycles of abrasion and wetting/drying. These laboratory test cycles can be related to in situ environmental stresses by estimating the number of times the rip rap will be flooded using site-specific hydrology and flood recurrence statistics (slake abrasion) and the number of times per year that rip rap and rock mulch will be exposed to wetting and drying. The latter estimate has the greatest uncertainty. These tests do not take into account long-term chemical weathering that will affect the rocks during the containment period and how that might decrease their resistance to abrasion.

Demonstration Phase - The "best" rocks from the reconnaissance phase, as determined by their relative durability and cost (a function of durability as it affects the size of rock needed and of haul distance to the site) must be shown quantitatively to be resistant to long-term chemical weathering. The procedure that we will recommend in the final stabilization handbook is to revisit the field at localities chosen specifically for gathering samples of the candidate lithologies that have been exposed to weathering for different and measurable lengths of time. Using detailed local stratigraphy, geomorphic determinations of relative weathering durations, and absolute determinations of age where possible (e.g., by radiocarbon dating, dendrochronology, leaching of desert varnish, etc.) should provide a suite of samples of the same rock that can provide a measure of the variability in resistance to wet abrasion as a function of weathering duration.

Subtask H. Impoundment Slope Stability and Settlement

The initial draft of the subtask report was completed during March and is currently undergoing review and editing.

PROJECTED WORK FOR NEXT QUARTER

The final reports for subtasks A, B and C (combined), E, G, and H are to be completed. Work on subtask F (Watershed Model Evaluation) will continue with the testing of mathematical models. Work will commence on the riprap engineering handbook.

REPORTS ISSUED DURING THE QUARTER

Beedlow, P. A. 1984. Designing Vegetation Covers for Long-Term Stabilization of Uranium Mill Tailings. NUREG/CR-3674 (PNL-4986) U. S. Nuclear Regulatory Commission, Washington, D.C.

CONFERENCE PRESENTATIONS

Three technical papers were presented at the Sixth Symposium, Management of Uranium Mill Tailings, Low-Level Waste and Hazardous Waste, held at Ft. Collins, CO, February 1-3, 1984.

1) Walters, W. H. and R. L. Skaggs. "Hydrologic Considerations for Rock Riprap Protection of Uranium Tailings Impoundments."

2) Beedlow, P. A. and M. C. McShane. "The Suitability of Vegetation for Erosion Control on Uranium Mill Tailings: A Regional Analysis."

3) Beedlow, P. A. and D. W. Carlisle. "Long-Term Stabilization of Uranium Mill Tailings: Effects of Rock Material on Vegetation and Soil Moisture."

TASK. 2. INTERIM STABILIZATION OF TAILINGS

Task Project Manager: J. N. Hartley
Principal Investigator: M. R. Elmore

FIN B-2370 (Task 2)

OBJECTIVE

The overall objective of this task is to assess the effectiveness, durability, and practicability of interim stabilization techniques and strategies for the suppression of fugitive dust from exposed tailings surfaces, under a full range of site and environmental conditions.

ACCOMPLISHMENTS DURING PAST QUARTER

A paper, "Fugitive Dust Control at Uranium Mill Tailings Piles," was presented at the Sixth Annual Symposium on Mill Tailings Management, Low-Level Waste and Hazardous Waste, Colorado State University at Fort Collins, Colorado on February 1-3, 1984. Also a journal article, "A Review of Fugitive Dust Control Techniques for Uranium Mill Tailings Impoundments," was submitted for publication in Nuclear Safety. The report, Laboratory Testing of Chemical Stabilizers for Control of Fugitive Dust Emissions from Uranium Mill Tailings was completed and submitted to NRC for printing.

The PNL windscreen field test is being monitored and data analyzed on the effectiveness of the three types of screens. Data are accumulated by a portable data logger and collected for analysis on a weekly basis. Preliminary results are showing an area of expected wind velocity reduction downwind of the screens. Comparisons of the screens will be made when sufficient data are available.

PROJECTED WORK FOR NEXT QUARTER

Work will continue on the evaluation of windscreens at the PNL field test site. Another type of windscreen has been identified and may be added to the present field test. Also, a monitoring trip to the Wyoming test site is being tentatively scheduled for the next quarter.

TASK 3. TAILINGS DEWATERING TECHNIQUES

Task Project Manager: M. J. Fayer
Principal Investigators: M. J. Fayer
T. J. McKeon
S. W. Tyler

FIN B-2370 (Task 3)

OBJECTIVE

In this task we determine the most effective approaches to dewatering uranium mill tailings that have been slurried into pits. The primary goals are to get an earlier placement of radon suppression covers and to minimize the release of contaminants to the environment by reducing the seepage into ground water. Special attention is given to evaluating the classical vertical consolidation theory as a worst-case estimator of settlement during the dewatering of tailings. Improved analysis techniques will be implemented and we will evaluate the test measurement techniques for obtaining the material properties (i.e., void ratio, degree of saturation, and hydraulic conductivity) as functions of the stress-state parameters. We will use this analysis capability to assess various disposal strategies.

ACCOMPLISHMENTS DURING PAST QUARTER

Subtask A. Evaluation of Linear Consolidation Theory

The work on this subtask was completed in FY82.

Subtask B. Measurement and Representation of Material Characteristics

The objective of this subtask is to obtain the necessary saturation, hydraulic conductivity and void ratio data that will allow us to simulate fluid flow and consolidation in uranium mill tailings piles. We will also develop methods for interpreting the lab data.

The measurements of saturated Grand Junction mill tailing sands, slimes, and a sand/slime mix have been completed. Measurements of unsaturated mill tailing slimes are continuing. Some of the earlier measured data are suspect, however, following the discovery of two sources of experimental error. One source was easily corrected but the other required slight modification of the equipment (the Rowe consolidometer) to eliminate the error.

Because of the nature of the partially saturated experiment, it will take at least six months to complete the analysis for any one material. Because slimes are expected to consolidate the most, attention will be focused on measuring the partially saturated consolidation of slimes first. Time may preclude the partially saturated testing of all three materials.

Subtask C. Computer Code Modification Testing and Documentation

The objective of this subtask is to change the existing consolidation model in the TRUST code to the two stress-state model. This change primarily involves making void ratio, saturation, and hydraulic conductivity functions of both the load at a point in the tailings pile and the local capillary pressure. It was originally thought that this representation of the material properties would only be used in the partially saturated region, and that classical consolidation theory would still be used in the saturated region. Simulations of slime consolidation, however, have shown two problems with this approach.

The first problem occurs when a layer of tailings becomes partially saturated. At that point, the modelled void ratio goes from being described by a compression index in the saturated zone to being described by stress-state parameters in the partially saturated zone. The change in void ratio is discontinuous at this point and presents problems for the model calculations.

The second problem occurs with the use of one value for the compression index in the saturated zone. In many instances, a single compression index does not always fit the data over the whole range of stresses to be encountered, especially at low stresses. Thus, void ratios calculated at low stresses can be erroneously high and lead to inflated estimates of drainage, total settlement, and time to complete settlement.

The solution to both problems is to extend the use of stress-state parameters into the saturated zone; i.e., use a non-linear relation that fits the measured void ratio versus effective stress data rather than approximating the relation linearly as is done in classical consolidation theory. Thus, changes in void ratio are continuous at all times and saturated void ratio changes are consistent with measured data at all stresses.

Subtask D. Evaluation of the Interactions Between Consolidation and Fluid Flow

Work on this subtask was delayed until enough data in the partially saturated region were available.

Subtask E. Tailings Dewatering Test Cases

No work on this subtask was scheduled for this report period.

PROJECTED WORK FOR NEXT QUARTER

During the third quarter of FY84, work will continue on obtaining the partially saturated property data for slimes from the Grand Junction mill tailings pile. Comparison studies to be conducted under Subtask D will be performed. Until all the experiments are finished, the comparison studies will use the slimes data generated to date, supplemented with some data published by Sherry (1980). We expect the slimes to consolidate more than other materials that will be measured (i.e., sands and a sand/slime mix).

Therefore, the effects of coupling and decoupling the flow and consolidation are expected to be most significant for the slimes. Documentation of the variably saturated flow and consolidation computer model TRUST-II will be continued. A first draft of the TRUST-II document will be prepared.

REFERENCES

Sherry, G.P. 1980. Constitutive-Relationships for Unsaturated Uranium Mill Tailings. MS Thesis. Colorado State University, Ft. Collins, Colorado.

TASK 4. TAILINGS NEUTRALIZATION AND OTHER ALTERNATIVES FOR IMMOBILIZING TOXIC MATERIALS IN TAILINGS

Task Project Manager: B. E. Opitz
Principal Investigators: B. E. Opitz
R. J. Serne
Support Investigators: M. E. Dodson
R. L. Erikson
M. J. Mason
D. R. Sherwood

FIN B-2370 (Task 4)

OBJECTIVE

In this task we assess the effectiveness, benefits, and costs of treating acidic tailings and tailings solution to reduce the potential leaching of toxic elements, radionuclides and macro ions from a tailings impoundment.

ACCOMPLISHMENTS DURING PAST QUARTER

Subtask A. Neutralization Methods Selection

This subtask was completed in FY83 when we completed and submitted the literature review on amelioration techniques. However, throughout the remainder of the project we will keep abreast of recent developments in applicable treatment techniques.

Subtask B. Laboratory Analysis

Laboratory experiments evaluating possible contaminant/carbonate complexation were initiated during this quarter. The tests are designed to evaluate the dissolution of previously precipitated contaminants when contacted with solutions high in carbonate content. Conditions of this nature could arise if acidic tailings solution were neutralized with only limestone or if uranium mill waste was disposed of in a waste impoundment surrounded by highly calcareous sediment. Results of earlier laboratory progress reports indicated that constituents such as Co, Mn, Mo, and perhaps U may redissolve and become mobile when high carbonate contents are present in solution. Preliminary results of these experiments indicate that increased concentrations of the above mentioned elements was not only a function of carbonate content but also of solution pH, most notably pH 8.0 and above.

Radium attenuation experiments were performed on mill tailings during this quarter. Samples of tailings were treated with various combinations of lime, calcium sulfate, and barium chloride and then leached with a laboratory prepared groundwater. The column effluents from the leaching studies were then analyzed for cations, anions, and radium contents. Results of these experiments showed radium activity reductions of greater than four orders of magnitude. Leachable radium activities in the untreated tailings were approximately

3500 pCi/L and were reduced to less than 5 pCi/L in the first effluents collected and remained at that level through the course of the leaching experiments. Further studies are needed to evaluate the long term effects of these treatment schemes.

Subtask C. Field Tests

A field demonstration plan entitled "Field Demonstration Plan for Verification of Uranium Mill Tailings Neutralization Techniques," was submitted during the second quarter of FY 83. The plan outlines three alternatives for field testing the most promising and cost-effective current neutralization technique.

REPORTS ISSUED DURING THE QUARTER

The paper entitled "Neutralizing Barriers for Reducing Contaminant Migration from an Uranium Disposal Pond," was presented at the 6th Annual Symposium on Management of Uranium Mill Tailings, Low-Level Waste, and Hazardous Waste held in Ft. Collins, Colorado, 3 February 1984.

PROJECTED WORK FOR NEXT QUARTER

Laboratory work has been completed; remaining efforts will be to prepare the final report.

TASK 5. EVALUATION OF SEEPAGE AND LEACHATE TRANSPORT FROM TAILINGS DISPOSAL FACILITIES

Task Project Manager: R. W. Nelson
Assistant Task Manager: T. J. McKeon
Principal Investigators: A. E. Reisenauer
C. J. Hostetler

FIN B-2370 (Task 5)

OBJECTIVE

In this task we evaluate the environmental consequences that may result from the disposal of uranium mill tailings below the water table. Two methods for the disposal of tailings below the water table are examined. The first involves disposal in excavated pits where some or all of the tailings are in contact with the regional ground-water flow and subject to leching by the ground water. The second method being currently studied is the disposal of tailings in deep underground mine stopes below the water table.

ACCOMPLISHMENTS DURING PAST QUARTER

The method developed during the first quarter of this year was used for evaluating the environmental consequences resulting from the disposal of uranium mill tailings in deep underground mine stopes. The evaluation method used is described in NUREG/CR-3560, Evaluation Methods for the Consequences of Below Water Table Mine Disposal of Uranium Mill Tailings, (McKeon and Nelson 1982).

Our purpose was to identify those situations where the most serious environmental consequences may be expected to occur. In particular, the consequences were investigated for the various combinations of changes in systems parameters. Those parameters studied include: the contrast in hydraulic conductivity between the tailings and the aquifer, the orientation of the mine stope with respect to the regional gradient, the size of the stope and its length, the regional groundwater gradient, the pumping rate from the downgradient well, and the location of the well.

The evaluations to date indicate three parameters are of major importance to the environmental consequence since they primarily control the leachate plume shape downgradient from the tailings filled stope. The angle between the regional gradient and the longitudinal axis of the stope is of greatest significance followed by the contrast of hydraulic conductivities, i.e., the ratio of tailings conductivity to the aquifer hydraulic conductivity. The third parameter in decreasing order of importance is the ratio of stope length to stope diameter. These three system parameters are of primary importance since they have major control over the shape of the leachate plume that develops downgradient from the stope.

When the stope is oriented perpendicular to the regional gradient, i.e., when the orientation angle is $\theta = 90°$, then the leachate plume is the widest possible for a given length of stope, the plume is moderately short in length, and it is shallower in vertical thickness than in any other orientation in the regional ground-water flow system. As the orientation angle diminishes from 90° toward 45°, there is some reduction in plume width, though not excessive, and a mildly increasing depth of the contaminated plume. Also, the plume length increases somewhat and greater variations in plume length across the width of the plume are noted.

The larger changes in leachate plume configuration occur as the orientation angle reduces further from 45° toward 0°, with ever increasing changes occurring as the length of the stope becomes more aligned with the direction of flow, i.e, θ approaches 0°. At zero degrees the width of the plume has reduced significantly and the depth has increased somewhat until they are now equal, and the plume length has grown very large. Under these aligned conditions, the width and depth of the plume is controlled almost exclusively by the contrast or the ratio of the tailings conductivity, to that of the regional aquifer hydraulic conductivity. The length of the contaminated plume depends almost exclusively on the length of the stope containing tailings.

Results to present indicate the three primary parameters controlling the plume shape can assume values so that the downgradient pumping well can be an inappropriate measure of the environmental consequences. For example, when the hydraulic conductivity of the aquifer is less than about 100 meters per year (3×10^{-4} cm/sec) and the tailings permeability is not more than 5 times greater than that of the aquifer, then not enough water can be pumped from the well in the worst case assumption that the well is perforated only over the depth of the leachate plume. In such systems of moderate to low hydraulic conductivity, the very small quantity of water pumped from the well is a direct result of the worst case assumption. In actual practice when seeking a water supply, perforations are not restricted to the depth of some contamination plume as the worst case analysis necessarily requires. However, if the well were perforated over greater lengths, then significant dilution would result from the uncontaminated water entering those perforations that are above and below the contamination plume. The method we have developed can evaluate such cases of greater dilution, but they generally do not give the worst case concentrations reaching the biosphere. There are also combinations of the stope length ratio and the angle between the gradient and the longest axis of the stope, where the width of the cone of depression for the well is less than the leachate plume width. Under such conditions the concentration in the well is not a sufficient measure of the complete environmental consequence.

Based upon the above findings, the emphasis for determining the consequences to the biosphere is being reoriented somewhat. The approach now being recommended involves using the model to determine the overall plume shape. This includes the maximum plume width and its vertical thickness. With the perforated length in the well set equal to the plume thickness, and given the desired well discharge, then the maximum width of the cone of depression for the well can be calculated. If the cone of depression of the well is less than the plume width, then the well provides a realistic assessment of the environmental consequences. However, if the width of the well cone of depression is greater than the plume width, then likely the down-gradient well should not be the sole basis for evaluating the consequences of the disposal of uranium tailings in deep mine stopes and tunnels.

The consequence analysis discussed above incorporates only one of two geochemical interactions, namely the leaching of the tailings by ground water. The others, interactions between the leachate and the porous medium of the aquifer after the leachate has left the mine stope, are ignored. The model developed for the consequence analysis is structured such that it may easily be coupled with the results of a geochemical code evaluating the interactions between the leachate and the porous medium.

Laboratory experiments studying the leaching of tailings by natural ground waters and the interactions of acidic leachate with natural aquifer materials are being conducted as part of Task 4 of this research, B-2370, and also under the NRC-sponsored project B-2292. Results from the leaching studies of mill tailings have been summarized and are ready for incorporation into the model used to predict the consequences of tailings disposal in mine stopes.

The leachate breakthrough data from experimental columns of coarser aquifer material conducted during the last year by other cooperating researchers have been assembled and were being used as a basis to test the initial geochemical coupled-transport simulation model. Specifically, the tailings solution concentrations input to the experimental columns along with the characteristics of the coarser aquifer material in the columns would be used to calculate the expected leachate breakthrough curve. The testing was started during the quarter using the initial code developed last year at PNL. Difficulties were found in the existing code. Specifically, the activity coefficients were not updated with changes in ionic strength except at the start of using the code. This was unrealistic in our system where the pH is changing by 5 to 6 orders of magnitude. The input solution chemical make up had to be constant and could not be time dependent as would be required to analyze the interactions of tailings leachate with aquifer materials. When these limitations were realized, work was started to remedy them. The modifications are being jointly funded by Tasks 4 and 5 of B-2292 and B-2379 with completion scheduled for mid-April. Once these modifications are made, the testing will resume.

The progress both in chemical interaction and fluid flow toward better understanding the performance of the various effects in the tailings stope disposal scheme has been significant and is leading to what now appears to be a very useful assessment procedure. The insights and understanding being obtained are demonstrating some very useful evaluation methods that will be simple to use. Though the assessment and analysis have been rather involved to gain the understanding of interactions, are now deriving simpler guidelines and evaluation tools.

PROJECTED WORK FOR NEXT QUARTER

The modification of the initial coupled transport and geochemical code will be completed by mid-April and the testing of the code using experimental data will then resume. It is hoped that these results will be sufficiently accurate to enable use of the code to obtain the tailings-leachate/porous aquifer chemical interactions. Such interactions will then be incorporated into the fluid flow analysis.

Work will also continue with the fluid flow evaluations to provide simplified useful methods and understanding to aid in the design and evaluation of deep mine disposal alternatives for uranium tailings. It now appears that the fluid flow assessment may be considerably simplified, perhaps to enable initial analysis at the hand-held calculator level of sophistication.

REFERENCES

McKeon, T. J. and R. W. Nelson. 1983. Evaluation Methods for the Consequences of Below Water Table Mine Disposal of Uranium Mill Tailings. NUREG/CR-3560, Nuclear Regulatory Commission, Washington, D.C.

Felmy, A. R., A. E. Reisenauer, Z. M. Zachara, and G. W. Gee. 1984. MININR: Geochemical Computer Program for Inclusion in-Water Flow Models--An Application Study. PNL-4921. Pacific Northwest Laboratory, Richland, Washington.

TASK 6. EFFLUENT AND ENVIRONMENTAL MONITORING METHODS AND EQUIPMENT AND INSTRUMENT TESTING

Project Manager: N. A. Wogman
Principal Investigators: P. O. Jackson
E. A. Lepel
K. B. Olsen
V. W. Thomas
W. C. Weimer
J. A. Young

FIN B-2370 (Task 6)

OBJECTIVE

In this task we assess measuring devices, techniques, and procedures for verifying if site and structure decontamination is acceptable. We develop needed monitoring methods and equipment, including methods to assess the release of radioactive and toxic materials from various facility components. From these assessments, we can provide guidance on monitoring requirements and strategies to evaluate compliance with uranium mill and tailings standards (e.g., PL-95-604, 40 CFR 192, 40 CFR 190). The work in this task is distributed among several subtasks identified below.

ACCOMPLISHMENTS DURING PAST QUARTER

Subtask A. Field Comparison of Radon Progeny Measurement Techniques and Critical Evaluation of Long-Term Radon-Daughter Measurements and Needed Improvements

Objective

In this subtask we ascertain whether a satisfactory short-term measurement technique is available for the determination of annual-average radon-daughter concentrations in buildings. We are to determine which, if any, of the available long-term radon and radon-daughter measuring devices is the most accurate and cost-effective.

Accomplishments During Past Quarter

Preparation of a letter status report continued.

Projected Work for Next Quarter

The letter status report will be completed.

Subtask B. Yellowcake Emission From Ventilation Exhausts

Objective

In this subtask we are to evaluate current methods for monitoring yellowcake and to recommend a new, revised, or improved method for monitoring yellowcake emissions from ventilation exhausts at uranium mill sites.

Accomplishments During Past Quarter

Preparation of a letter status report continued.

Projected Work for Next Quarter

The status report will be completed.

Subtask D. Critical Evaluation of Measurement Techniques for Uranium ^{226}Ra, ^{230}Th, and ^{210}Pb in Effluents and Environmental Materials and Recommended Improvements

Objective

In this subtask we compare the sensitivity of existing analytical procedures for quantifying U, ^{226}Ra, ^{230}Th, and ^{210}Pb in effluents and environmental materials. The individual sensitivities will be compared with regulatory standards. Where improvements are required, existing techniques will be extended, if possible, or new techniques will be developed to produce methods which are sensitive enough to address the regulatory standards. The final report for this subtask will be a set of analysis methods to be used to measure the radioisotopes of interest in both effluents and environmental materials.

Accomplishments During Past Quarter

A status report, "Measurement Techniques for Uranium, ^{226}Ra, ^{230}Th, and ^{210}Pb in Effluents and Environmental Materials Associated with Uranium Recovery," was issued.

Projected Work for Next Quarter

None.

Subtask E. Review Well-Logging Techniques

Objective

In this subtask we are to 1) evaluate and compare well-logging techniques with core-sample analytical results for the measurement of uranium and its progeny in uranium tailings, 2) determine the effects of interfaces (high natural concentrations of uranium decay-chain products and variations of ^{40}K concentrations) on the response of the well-logging instruments, and

3) recommend an acceptable procedure which can be applied to present and future uranium mill tailings remedial-action programs.

Accomplishments During Past Quarter

The report, "Borehole Logging During Engineering Assessment for Remedial Action--Recommended Procedures and Equipment," was reviewed, and readied for submittal to Clearance.

Projected Work for Next Quarter

We will prepare a camera-ready copy of the report mentioned above and submit it to NRC once it is cleared.

Subtask F. Evaluate and Recommend Sampling Strategies for Temporal and Spatial Evaluation of Contamination

Objective

In this subtask we are to develop guidelines to help detect and evaluate the windborne-radioactive contamination that has been spread from uranium tailings piles and which may extend up to several kilometers. Our guidelines will define the number, distribution, and timing of samples necessary to detect the spread of contamination with reasonable certainty, while keeping the cost and size of the detection effort at a minimum.

Accomplishments During Past Quarter

No activity.

Projected Work for Next Quarter

None.

Subtask G. Sampling Techniques for Detecting Contamination of Surface and Ground Water by Tailings Leachate

Objective

In this subtask we are to evaluate the existing standard techniques for measuring levels of contaminants in natural waters to determine their applicability at uranium recovery sites. Known dissolved contaminants that are potential hazards to water quality will be considered in this study. These include some of the radioactive uranium-daughter products (in particular thorium and radium) and stable elements Se, As, Sb, Zn, Ni, Cr, and Cu. An additional objective is to propose a spatially-oriented monitoring protocol for ground water that will be generally useful for uranium recovery sites.

Accomplishments During Past Quarter

No activity.

Projected Work for Next Quarter

None.

Subtask I. Radon Flux Comparisons with ^{226}Ra Concentrations in Surface and Subsurface Soils

Objective

We will examine the degree of correlation that exists between radon fluxes and ^{226}Ra concentrations in surface and subsurface soil to determine whether average radon fluxes can be predicted from measurements of ^{226}Ra concentrations in soil and whether detection and delineation of subsurface deposits of ^{226}Ra can be enhanced by the use of radon-flux measurements at the soil surface.

Accomplishments During Past Quarter

The status report Comparison of Radon Fluxes with Gamma-Ray-Exposure Rates and Soil ^{226}Ra Concentrations, was completed.

Projected Work for Next Quarter

None.

REPORTS ISSUED DURING THE QUARTER

Young, J. A. and V. W. Thomas. 1984. Comparison of Radon Fluxes With Gamma Ray Exposure Rates and Soil ^{226}Ra Concentrations, NUREG/CR-5016, PNL-3677, Nuclear Regulatory Commission, Washington, D.C.

TASK 7. TECHNICAL PROGRAM MANAGEMENT

Project Manager: M. G. Foley
Principal Investigators: M. G. Foley
G. W. Gee

FIN B-2370 (Task 7)

OBJECTIVE

This task provides the program management function of all the uranium recovery waste management studies contracted to PNL from the Waste Management Branch, NRC-RES. The principal functions of Program Management are to coordinate the efforts of scientists and engineers assigned to tasks, prepare and issue quarterly technical and administrative reports, maintain an awareness of uranium waste research by others, schedule and arrange local and external research reviews, and carry out related management functions.

ACCOMPLISHMENTS DURING THE PAST QUARTER

The program manager is the principal point of contact between the PNL technical team and NRC-RES project and task managers, and must ensure that PNL work addresses the research needs of the NRC; that NRC is kept informed of PNL progress as well as final products; and that all deliverables provided to NRC have been properly reviewed. Specific activities over the past quarter to accomplish this task are listed below.

- Made arrangements for an NRC review of the uranium recovery research program to be held at PNL in April.
- Reviewed the drafts and final documents of the reports produced by the uranium recovery program (listed elsewhere in this report).
- Prepared and submitted 3 monthly administrative reports.

International Activities

Canadian Mill Tailings Program

M. G. Foley met with Mr. Vic Haw, director of the Canadian National Uranium Tailings Program, during the February, 1984 Uranium Mill Tailings Symposium in Fort Collins, Colorado, as a follow-up to our visit with him in Ottawa in September.

Australian Mill Tailings Program

Ms. Kaye Hart of the Australian Atomic Energy Commission met with program staff at PNL on 6 and 7 February to discuss technical aspects of the U. S. and Australian approaches to disposal of uranium mill tailings.

IAEA Participation

Dr. G. W. Gee attended the Coordinated Research Program meeting of the International Atomic Energy Agency (IAEA) in Brazil in February, 1984. The IAEA is covering the travel costs for Dr. Gee to attend this meeting. The NRC has a no cost-research agreement with the IAEA, allowing information exchange on research related to contaminant migration from uranium mill tailings. The workshop type meeting provided exchange of current research from Argentina, Australia, Brazil, Canada, Czechoslovakia, India, Japan, and the U.S.

PROJECTED WORK FOR NEXT QUARTER

In addition to managing the technical and budgetary schedules, and submitting periodic administrative and technical progress reports, the program manager will ensure that:

- Preparations for closeout of the program are begun in good order.
- Continuing reviews of PNL work by internal and external peer reviewers and by NRC will be arranged and conducted.

REPORTS ISSUED DURING THE QUARTER

Foley, M. G. et al. 1984. Uranium Recovery Research Sponsored by the Nuclear Regulatory Commission at Pacific Northwest Laboratory: Annual Progress Report, October 1983 to December 1983. (PNL-5015-1), Pacific Northwest Laboratory, Richland, Washington.

ATTENUATION OF RADON EMISSION FROM URANIUM MILL TAILINGS

Project Manager: N. A. Wogman
Principal Investigator: D. R. Kalkwarf
Support Investigators: V. C. Rogers and K. K. Nielson (Rogers and Associates Engineering Corporation) RAE

FIN B-2269

OBJECTIVE

In this project we will define uniform methods for determining the thickness of soil cover material needed to meet radon flux limits (as specified in connection with the final GEIS on uranium milling), and design methods for measuring the effectiveness of the soil cover after it is emplaced. These methods will be used to develop a NRC regulatory guide.

ACCOMPLISHMENTS DURING PAST QUARTER

Task 8. Prepare a Handbook for Determining Radon Attenuation Cover Soils (RAE)

Objective

We are to prepare a handbook to guide engineers in the covering of uranium mill tailings piles with soil in order to reduce radon emission to prescribed levels.

Accomplishments During Report Period

A camera-ready copy of the document, Radon Attenuation Handbook for Uranium Mill Tailings Cover Design, NUREG/CR-3533, was delivered to NRC.

Task. 10. Validate Calculated Radon Attenuation Values for Soils Covering Large Areas of Tailings

Objective

We are to validate radon attenuation values, calculated by methods described in the revised engineering handbook, for large areas of tailings.

Accomplishments During Report Period

Methods described in the final version of the Handbook were found to generate conservative estimates for the thickness of earthen covers needed to attenuate radon flux to prescribed levels at the UMTRAP site, Grand Junction, Colorado. A document describing these results is being completed, utilizing equations and methods described in "the Handbook".

Task 12. Prepare a Regulatory Guide for Determining Radon Attenuation by Cover Soils (RAE)

Objective

We are to prepare a document in the format of an NRC Regulatory Guide which specifies acceptable methods for covering uranium-mill tailings with soil to reduce radon flux to within prescribed limits.

Accomplishments During Report Period

A camera-ready copy of this document was delivered to NRC.

PROJECTED WORK FOR NEXT QUARTER

This project was scheduled for completion in CY-1983. A camera-ready copy of a topical report describing tests of the validity of handbook methods using data collected at the Grand Junction site will be delivered to NRC as soon as peer review and clearance is obtained.

REPORTS ISSUED DURING THIS QUARTER

Kalkwarf, D. R. and W. B. Silker. 1984. "Diffusion of Radon in Candidate Soils for Covering Uranium-Mill Tailings". In Management of Uranium Mill Tailings, Low-Level Waste and Hazardous Waste; Proceedings of the Sixth Symposium on Uranium Mill Tailings Management, Colorado State University, Ft. Collins, Colorado.

Nielson, K. K. and V. C. Rogers. 1984. Calculation of Radon Flux Reduction by Uranium Mill Tailings Covers: A Report for Preparation of a Regulatory Guide.

Rogers, V. C. and K. K. Nielson. 1984. Radon Attenuation Handbook for Uranium Mill Tailings Cover Design, NUREG/CR-3533. U. S. Nuclear Regulatory Commission, Washington, D. C.

ASSESSMENT OF LEACHATE MOVEMENT FROM URANIUM MILL TAILINGS

Project Manager: S. R. Peterson
Principal Investigators: R. J. Serne
W. J. Martin
S. R. Peterson
M. E. Dodson
Support Investigators: M. J. Mason
G. W. Gee
B. E. Opitz
R. L. Erikson

FIN B-2292

OBJECTIVE

Within this program we will develop experimental and computer modeling tools to assess the long-term environmental impact of leachate movement from uranium mill tailings and commercial low-level waste.

ACCOMPLISHMENTS DURING REPORT PERIOD

Task 1. Project Management

As of the first of February this program is no longer funded under Uranium Recovery. Funds have been made available through Low-Level Waste to fund this work through FY84. The NRC technical monitor will continue to be George Birchard.

The paper prepared for future inclusion in a special NUREG document was reformatted. This paper is entitled, "Interactions of Acidic Solutions with Sediments: A Case Study." Minor reformatting was necessary to bring our paper into compliance with the style of the special NUREG document.

Task 2. Laboratory Analysis and Experiments

Two types of experiments are currently being conducted. In the first type, leaching experiments were designed with various column lengths and diameters. Leachate flow through the columns was from bottom to top to ensure complete column saturation. Synthetic ground water was used as a leachate. Two of the columns had the same length to diameter ratio (1 to 1) so that the only factor varying between the two columns was the volume. Other columns were constructed that had the same diameters but different lengths. The synthetic ground water was then passed through these columns after packing them with uranium mill tailings.

There appear to be no effects on the effluent concentrations of the leaching columns due to differences in column lengths (residence time) or column volumes (scaling). The results of these experiments provide a foundation for using column experiments as a technique in leaching uranium mill tailings. These results may then be used in predicting long- and short-term movement of contaminants from tailings impoundments because of leaching due to migrating ground waters or rainfall.

The objective of the second study is to evaluate the effects of residence time when acidic uranium mill tailings solutions contact coarse sediments. Material from a mill shaftwall was used in the residence time experiments. Particle size determinations indicate that the sediment was comprised of approximately 80% sand sized particles. Furthermore, the calcium carbonate content was determined to be less than 0.5% by weight. This indicates a low buffering capacity.

Samples of the shaftwall material were compacted into flow-through columns and tailings solution was pumped through the sediment at various flow rates to allow sediment/solution residence times to vary.

The column residence times were calculated based on the length of time one pore volume of solution resided in the column. The duration of pore volume contact ranged from 20 hours to 140 hours per pore volume. Using the 20 hour per pore volume duration as the base residence time (1:1), the remaining columns varied from 2:1 (40 hours per pore volume) up to 7:1 (140 hours per pore volume).

Preliminary results of the column effluent characterization indicate that for nonreactive species, (e.g., Cl), the duration of the tailings solution/sediment interaction has no effect on the Cl breakthrough. For other micro ions such as Al, Ca, Fe, Si, and sulfate, the columns with the longer residence times seem to exhibit breakthrough at a later pore volume. Trace metal concentrations in the column effluents are currently being analyzed and the data should be available shortly. All conclusions drawn thus far with regard to these columns are very tentative. More definitive interpretations will be possible once all data are obtained.

Task 3. Geochemical Modeling

These coarse-sediment data will initially be evaluated using speciation and solubility modeling to determine potential solubility controls in the various columns. These results will be compared to the results obtained from modeling columns packed with clay. Most modeling predictions to date have considered flow through clay materials with relatively high adsorptive capacities. Modeling the coarse materials would allow us to delineate the contributions that adsorption makes to contaminant attenuation and to further substantiate our findings to date, i.e., macrocations are being attenuated by precipitation/dissolution reactions and trace metals by adsorption processes.

Furthermore, the modeling will allow us to explore the role that kinetis plays in these experiments. That is, are kinetics important in some of the precipitation/dissolution reactions that have been identified as controlling the concentrations of certain contaminants?

Field Study

Speciation and solubility modeling have been completed on the available well data at the evaporation pond near Riverton, Wyoming. Data were available from 17 wells. These data have been compiled in tables and a conceptual chemical model for the field site has been completed. This conceptual chemical model is currently being used, along with the MINTEQ computer code, to predict the concentrations of contaminants with distance from the tailings pond.

PROJECTED WORK FOR NEXT QUARTER

Continued analysis of the data obtained from the scaling and residence time experiments will continue. Additionally, the concentrations of contaminants in the effluent solutions from permeability cells packed with coarse overburden materials and leached with acidic tailings solution will be determined. These data will be modeled and compared to modeling results obtained when clay materials were leached with the same acidic solutions.

Additional characterization of sediments using techniques such as electron microprobe analysis, x-ray fluorescence and scanning electron midroscopy will be performed on the field samples that were collected at the evaporation pond. The additional characterization techniques would allow trace element profiles to be identified and provide insight into how and with what minerals and solid phases the trace elements tend to be associated.

The steps taken in the field modeling validation, and the steps yet to be taken, are: 1) Speciation/solubility modeling will be performed on the solutions obtained from the monitoring wells. 2) A conceptual chemical model of the system will be formulated. 3) Predictions will be made as to the variability of contaminant concentrations with distance based upon the chosen conceptual model and upon plug flow. 4) A coupled hydrogeochemical code will be used to predict contaminant concentrations with distance based upon the conceptual chemical model of the system and upon the available hydrologic data. 5) An attempt will be made to incorporate adsorption into the conceptual model of the system.

METHODS OF MINIMIZING GROUND-WATER CONTAMINATION
FROM IN-SITU LEACH URANIUM MINING

Project Manager: W. J. Deutsch
Principal Investigators: W. J. Deutsch
R. J. Serne
Support Investigators: W. J. Martin
D. R. Sherwood

FIN B-2379

OBJECTIVE

In this study we evaluate in-situ uranium mining operations and restoration procedures and provide information to help NRC establish regulatory guidelines for the environmentally sound operation and abandonment of these mines. We evaluate existing production/restoration procedures and assess the potential for deleterious hydrogeochemical alterations resulting from lixiviant excursions and ore-zone mining.

ACCOMPLISHMENTS DURING PAST QUARTER

Task 1. Project Management

During February the document Aquifer Restoration Techniques for In Situ Leach Uranium Mines NUREG/CR-3104, PNL-4583 was published and distributed by NRC.

Task 2. Site Characterization

Differences in the outcome of natural restoration column experiments using Texas and Wyoming sediments prompted us to further characterize these sediments. In the experiments (discussed in detail in the following section) we found that when Wyoming reduced sediment was used in the column, dissolved uranium was not removed from solution flowing through the column as was the case noted for experiments with Texas reduced sediment. Because pyrite in the sediment is a prime reducing agent for oxygen and U(VI), we felt that pyrite concentration variations in the sediment might account for the behavior observed in the experiments. We used a method developed and tested by Lord (1982) to determine the amount of pyrite in the sediment. This method consists of dissolution and removal of solids other than pyrite from the sediment followed by dissolution of pyrite and quantitative measurement of released iron. Our detection limit for pyrite was 0.004 weight percent of the sediment.

The results of the pyrite determinations for various types of Wyoming and Texas sediments are shown in Table 1. Except for the oxidized sediment (recovered from a zone up the hydrologic gradient from the roll front), all the Texas sediments have at least 0.4% pyrite. The reduced sediment used in the Texas natural restoration column experiments (similar to sample number A36) had approximately 3% pyrite. In contrast, the Wyoming sediments have less than 0.5% pyrite except for sample B20 of reduced sediment which has 1%

pyrite. Sample B2 was used in the Wyoming natural restoration column experiment and it contained only 0.05% pyrite. Consequently, there was approximately 60 times as much pyrite present in the sediment used in the Texas column experiments as there was in the Wyoming experiment. This may be the primary factor that influenced uranium concentration in the effluents. Additional experiments are being planned to better define the importance of pyrite and other iron sulfide minerals in the process of aquifer restoration and long-term contaminant control. These experiments are described in the following section.

TABLE 1. PYRITE CONTENT OF SEDIMENT (weight %)

Texas		Wyoming	
Reduced Sediment		Reduced Sediment	
A27A	1.5 wt. %	B2A	0.05
A27B	2.2	B2B	0.05
A36A	3.0	B20A	1.0
A36B	2.9	B20B	1.0
Ore Zone Sediment		Ore Zone Sediment	
A11A	1.2	B29A	0.46
A11B	1.2	B29B	0.42
A38A	1.2	B34A	0.32
A38B	1.2	B34B	0.30
Oxidized Sediment		Oxidized Sediment	
A18-215A	0.04	B47A	0.01
A18-215B	0.02	B47B	0.004
A18-216A	0.02	B51A	0.02
A18-216B	0.02	B51B	0.008
Leached Ore			
A1-225A	1.10		
A1-225B	0.95		
A1-226A	0.49		
A1-226B	0.40		

Task 3. Natural Restoration Processes

During December, 1983 a column experiment designed to evaluate the potential for natural restoration at the North Platte, Wyoming mine site was conducted. The experiment was performed in a manner similar to that of the Texas natural restoration experiment described in detail by Deutsch et al. 1983a. The objective of the experiment was to determine if the concentration of uranium in pregnant lixiviant would be lowered by contact of the solution with reduced sediment collected down the hydrologic gradient from the roll front. If uranium is removed from solution and immobilized in the sediment, then natural processes in the ore zone aquifer could be expected to enhance aquifer restoration and a better case for long-term stability of the system could be made.

Preliminary results of the Wyoming natural restoration column experiment show a significantly different response for dissolved uranium than was found for the Texas experiment. Figure 1 shows the concentration of uranium in the effluent from the two duplicate Wyoming columns and for the Texas column which had the same dimensions as the Wyoming columns. Uranium movement through the Wyoming columns was retarded by contact with the sediment but influent concentration (14 ppm) is approached after 8 pore volumes have flowed through the columns. This contrasts to the Texas case in which effluent uranium concentration increases initially but never reaches the influent concentration of 52 ppm. Uranium in the Texas column effluents decreases rapidly after reaching its peak, and after a little over 5 pore volumes the concentration is much less than 1 ppm for the subsequent 6 pore volumes of effluent. The difference in response of dissolved uranium for the Texas and Wyoming columns may be due to the amount of pyrite in the sediments. The Texas sediments contained 3 weight percent pyrite and the Wyoming sediment had 0.05 weight percent pyrite. If pyrite reacts sufficiently rapidly to affect the Eh of the solution flowing through the columns, it could establish conditions in which U(IV) minerals are stable. The relatively low solubility of these minerals would limit the amount of uranium in solution to values similar to those found in the Texas column effluents after 5 pore volumes of flow. The small amount of pyrite in the Wyoming sediments used in the column experiment may not be enough to lower the Eh sufficiently to affect the amount of dissolved uranium. As shown in Table 1, not all of the reduced Wyoming sediment has such a low pyrite content as that selected for the column experiment, but, in general, there appears to be less pyrite at the Wyoming site sampled that for the Texas site.

In order to further evaluate the possible importance of pyrite on the system, a new set of batch experiments with Texas and Wyoming sediments was started in February. The basic design of our batch experiment is described in our progress letter of November, 1983 (Deutsch et al. 1983b). For the current batch experiment some of the sediments were amended with pyrite and FeS-coated sand to compare with the naturally-occurring pyrite in the sediment. This experiment will evaluate the effect on solution composition of long-term (up to 3-1/2 months) storage of lixiviant and sediment. It will be completed in June, 1984.

Task 4. Induced Restoration Techniques

The majority of the data from the column experiments designed to study the effects of ground-water sweeping on aquifer restoration have been evaluated. In these experiments column containing leachate ore from the Texas mine were saturated with weak lixiviant (U = 3 ppm) that was spiked with the common leach-generated contaminants arsenic (4.8 ppm), selenium (4.1 ppm), and molybdenum (5.0 ppm). Spiked lixiviant was pumped through each column until more than 2 pore volumes had contacted the sediment. The column influent solution was then changed from lixiviant to ground water that had been collected at the Texas site. The ground water was pumped through the columns to simulate sweep-induced restoration of the aquifer. The entire experiment was conducted in an anoxic chamber to approximate the reduced oxygen fugacities of the ore-zone environment.

In the last quarterly report we discussed the column effluent chemistry which showed that uranium is mobile during ground-water sweeping and that arsenic, selenium and to a lesser extent molybdenum are removed from solution by interactions with the sediment. During this quarter we analyzed the sediments from the columns by x-ray fluorescence methods to corroborate the solution chemistry data and identify locations in the columns where preferrential removal or addition of these contaminants might be occurring.

Sediment chemistry data are given in Table 2 for: 1) the leached ore from the Texas mine that was used to pack the columns, 2) the homogenized sample of all of the column #1 sediment after the experiment (designated GS-1), and 3) three segments of column #2 sediment after the experiment (designated GS-2: 0-1, 5-6, 9.8-10.8 cm). The interval measurements for GS-2 are from the influent end of the column. As was found from the solution data, the arsenic, selenium, and molybdenum sediment analyses show that these elements were removed from the spiked solution and deposited in the leached ore. Arsenic and selenium show a similar distribution in column GS-2 with their highest concentration in the first segment of the column. Arsenic concentration is 4 times greater than its baseline value in the first centimeter of the column, but shows no enhancement over baseline at the end of the column. Selenium concentration ranges from nearly six parts per million in the first segment of the column to about 1.8 parts per million in the final segment. This profile is entirely due to selenium deposition from the spiked lixiviant because there was no detectable selenium in the original leached ore. The molybdenum distribution in column GS-2 shows that this element is relatively mobile compared to arsenic and selenium although there appears to be some increase in molybdenum concentration compared to baseline for the final segment of the column. In contrast to these three elements, uranium is actually leached from the sediment used in the sweeping experiment. The amount remaining in the sediment after the experiment is approximately 1/3 of that originally in the sediment. The trend of increasing uranium concentration with distance down the column for GS-2 is what would be expected if the oxidizing capacity of the solution entering the column is decreased by oxidation and dissolution of uraninite as the solution moves through the column.

The solution and sediment chemistry data for the ground-water sweeping experiment are consistent. They show that, if residual uranium ore is present in the leached zone at the completion of mining, restoration of the aquifer for uranium may be inhibited due to slow, long-term oxidation of the uranium minerals during ground-water sweeping. The mobility of other redox-sensitive contaminants such as arsenic, selenium, and molybdenum may be reduced significantly due to water/sediment interactions and this will enhance restoration for these elements.

TABLE 2. CONCENTRATIONS OF SELECTED CONSTITUENTS IN TEXAS LEACHED ORE

Column GS-1 Contacted Leached Ore, and Column GS-2 Contacted Leached Ore Segments (concentration units are parts per million)

Element	Leached Ore	GS-1	GS-2 0-1 cm	GS-2 5-6 cm	GS-2 9.8-10.8 cm
As	2.4	5.85	10.0	3.25	2.59
Se	<0.9	4.32	5.93	2.27	1.83
Mo	2.1	3.2	1.9	2.6	2.94
U	122	36.4	33.5	38.6	45.4

The subcontract with the Twin Cities Research Center (TCRC) of the Bureau of Mines has been continued this fiscal year. During the past quarter the TCRC began a mine operator survey of experience with surface treatment restoration equipment. The objective of the survey is to provide data to evaluate surface treatment equipment that has been used at in situ leach uranium mines to restore the ground water after leaching. The results of the survey will be available in April, 1984. The TCRC is also conducting a set of flow-through column experiments to evaluate ground-water sweeping and chemical addition restoration methods. Preliminary results from these experiments should be available in June, 1984.

PROJECTED WORK FOR NEXT QUARTER

The batch experiment with Texas and Wyoming sediments contacting lixiviant will continue into June, 1984. Solutions and sediments will be analyzed on a regular basis and compared to results of similar experiments conducted last fiscal year. We will continue laboratory studies of chemical reductants as aids to aquifer restoration, concentrating on sodium sulfide and sodium sulfite.

REPORTS PUBLISHED DURING THE QUARTER

Deutsch, W.J.et al. 1984. Aquifer Restoration Techniques for In Situ Leach Uranium Mines. NUREG/CR-3104, PNL-4583, Nuclear Regulatory Commission, Washington, D.C

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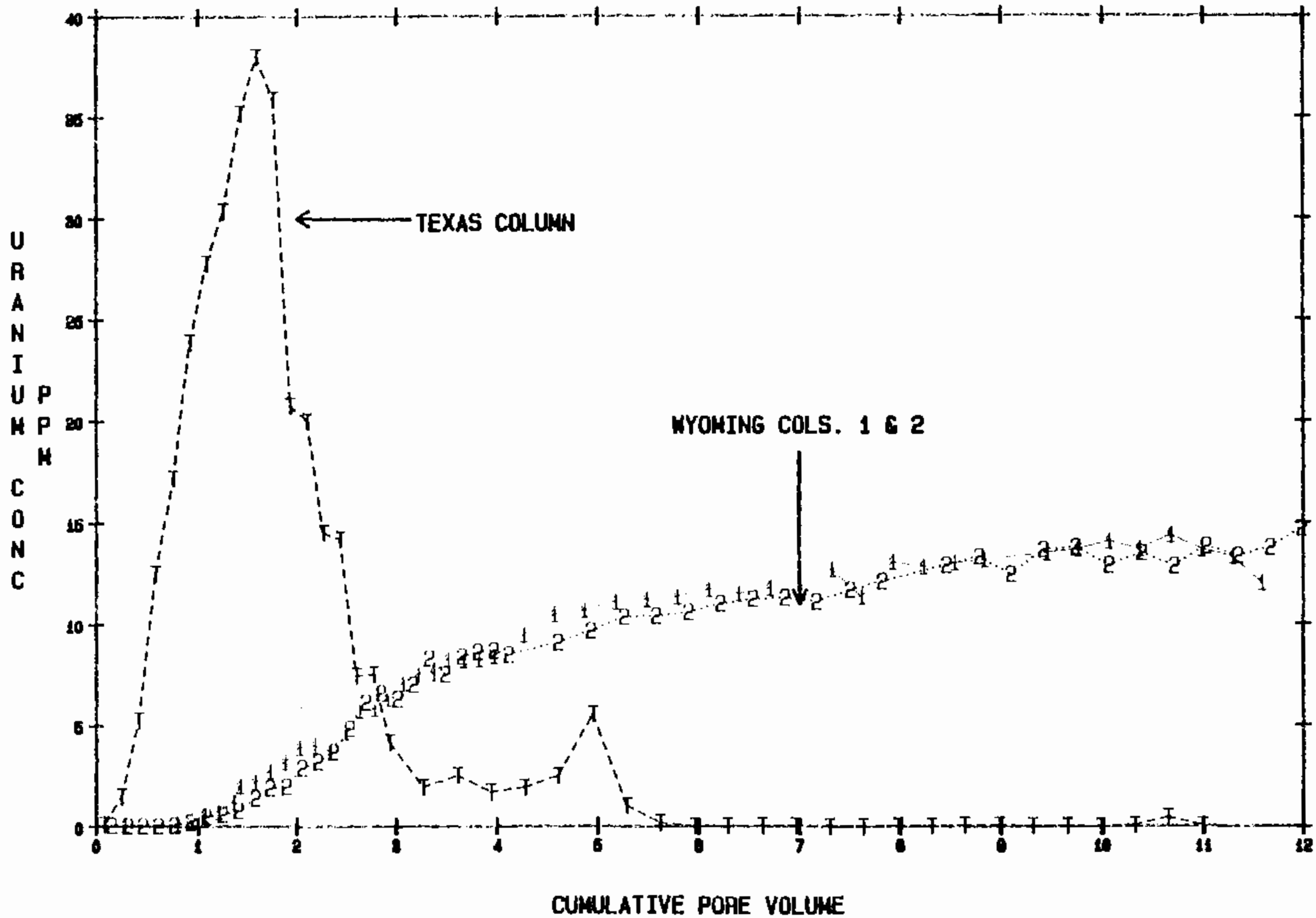
Deutsch, W.J., R.J. Serne, N.E. Bell and W.J. Martin. 1983a. Aquifer Restoration at In-Situ Leach Uranium Mines: Evidence for Natural Restoration Processes. NUREG/CR-3136, PNL-4506, Nuclear Regulatory Commission, Washington, D.C.

Deutsch, W.J., W. J. Martin, B. M. Sass, R. J. Serne. 1983b. Status of Field Work and Laboratory Experiments on Aquifer Restoration Following In Situ Leach Uranium Mining. Letter Progress Report. PNL-4924.

Lord, C.J.III. 1982. "A Selective and Precise Method for Pyrite Determination in Sedimentary Materials." J. Sed. Pet. 52:664-666.

FIGURE 1

URANIUM CONCENTRATION IN EFFLUENT FROM
TEXAS AND WYOMING NATURAL RESTORATION COLUMN EXPERIMENTS



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