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

**Quarterly Progress Report
April to June 1984**

August 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
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PACIFIC NORTHWEST LABORATORY
operated by
BATTELLE
for the
UNITED STATES DEPARTMENT OF ENERGY
under Contract DE-AC06-76RLO 1830

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

QUARTERLY PROGRESS REPORT
APRIL THROUGH JUNE 1984

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August 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

Work continued on the case study analysis of riprap design methods for the Grand Junction and Slickrock tailings impoundments. Sensitivity analyses were performed on the Dolores River hydraulic data to identify trends associated with variations in channel roughness (Mannings n), flow velocities, and design shear stress. The effects on the design stone size were evaluated. Initial test runs to evaluate the overland erosion model were completed. A journal article for Nuclear Safety was prepared and submitted to Oak Ridge National Laboratory.

- Interim Stabilization of Mill Tailings Piles

Monitoring of the Wyoming field test site was completed. The PNL field test monitoring will end in July. Results from these tests will be presented in the final report, which is being prepared. Nuclear Safety has accepted an article on this project for publication, and a paper is to be presented at an EPA-EPRI symposium on particulate control technology.

- Tailings Dewatering Techniques

The computer code which was developed for this task was originally designated as TRUST-II. This was not acceptable to the originator of the precursor code, TRUST, so the new code has been designated as TRUNC, for TRAnsient UNsaturated Consolidation.

Following the semi-annual review in April, we have changed the focus and due date of the report titled, TRUST-II: A Computer Program for Variably Saturated Flow in Deformable Media. The new title is, Settlement of Uranium Mill Tailings: A Comparison of Analysis Techniques and the report is due in September. Predictions made using empirical methods of settlement analysis are compared with predictions using TRUNC for both satu-rated and partially saturated conditions. We found that predictions of the amount of settlement were similar for both the empirical methods and TRUNC. However, only TRUNC was capable of predicting time of settlement under partially saturated conditions. In the case of a 12.2 m pile of slimes, TRUNC predicted 1500 days for settlement to be completed, which is about an order of magnitude greater than the predicted time for saturated settlement. Thus, partially saturated settlement may prove to be the controlling factor in determining when to place a cover on tailings piles. An appendix to the September report will contain a user's manual for the TRUNC code, as well as a section detailing the changes made to the TRUST code to arrive at the TRUNC formulation.

The measurement of material properties under partially saturated conditions has proven to be a time-consuming process. To allow time for more data to be gathered, the due date of the report, The Sampling and Measurement of Uranium Mill Tailings Consolidation Characteristics has been moved from September to December.

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

All ongoing laboratory experiments were completed during this quarter. Results of the contaminant/carbonate complexation studies and the tailings radium attenuation experiments are being compiled and will be documented in a laboratory progress report. Following the laboratory progress report, a final report covering the studies completed over the duration of this task will be submitted. At this time no further laboratory experiments are expected to be initiated before completing the final report for Task 4.

- Evaluation of Seepage and Leachate Transport From Tailings Disposal Facilities

The ground-water flow analysis and methodology for assessing leachate concentrations in the leachate seeping from uranium tailings disposed in deep mine stopes is essentially finished except for report preparation. Work is continuing on providing assessment methods for the chemical interactions of the tailings leachate with aquifer materials downgradient from the disposal stope.

The results for the ground-water flow and leachate plume downgradient from the tailings-filled mine stope are provided at two levels of detail. The first level provides the overall plume width, depth, and length as graphs of dimensionless parameters for direct use. The second, or more detailed, evaluation is provided by an interactive computer code that provides the concentration of leachate constituents in the discharge from a downgradient water supply well. The results of the ground-water flow analysis will be included in a report to be prepared during the next quarter entitled, Leachate Plume Migration Downgradient of Uranium Tailings Disposal in Deep Mine Stopes.

- Effluent and Environmental Monitoring Methods and Equipment and Instrument Testing

This task was not funded in FY84, and exhausted its funds carried over from FY83 in December. No work was performed in the last quarter except for preparation of final reports.

- Attenuation of Radon Emissions

A document describing the validation of methods presented in NUREG/CR-3533, Radon Attenuation Handbook for Uranium Mill Tailings Cover Design, is nearing completion and will be delivered to NRC next quarter.

- Assessment of Leachate Movement From Uranium Mill Tailings

This Project exhausted its uranium recovery funds carried over from FY83 in February 1984 and is now funded by Low-Level Waste.

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

Initial experiments to test the ability of sodium sulfide added to lixiviant to enhance aquifer restoration were completed. It was found that sulfide effectively reduces dissolved oxygen concentrations in solution. Removal of dissolved oxygen should limit the production of redox-sensitive trace metal contaminants (uranium, arsenic, selenium, and molybdenum) during restoration by recirculation methods. At sulfide concentrations of 330 mg/L the uranium concentration of the solution is also lowered suggesting the production of a U(IV) mineral. The addition of sodium sulfide raises the pH of the solution, which causes carbonate minerals to precipitate in an amount proportional to the rise in pH. This could have deleterious effects on restoration if the permeability of the aquifer is reduced appreciably.

A geochemical model of natural aquifer restoration is being developed using the data from a column experiment in which Wyoming sediment was contacted with synthetic lixiviant. The model will consider chemical processes such as oxidation/reduction, dissolution/precipitation, ion exchange, adsorption, and aqueous complexation. The computer code MINTEQ will be used to make the necessary calculations and provide modeling results to compare with column solution data. The successful development and validation of such a model will provide a method for simulating the effect of various restoration treatments on the natural system. It will be used to evaluate the chemical reductant experiments.

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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."

Uranium recovery research sponsored by the NRC 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

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

- (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

the projects endeavor 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 ground water in in situ extraction operations
- evaluate natural and induced restoration of ground water 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 April, 1984 through June, 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.

Mexico. In publication through the Committee on Onshore Energy Mineral Management Research sponsored by the National Research Council, Washington, D. C.

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 Investigators: 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. Other studies performed under this task will 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 slope stability.

ACCOMPLISHMENTS DURING PAST QUARTER

Subtask A. Technical Feasibility Study

Work continued on the sensitivity analysis of riprap design to the Dolores River hydraulic data for several potential flood events at the Slickrock, Colorado impoundment. The Mannings n roughness coefficient was varied by 10%, 25% and 50% for each of the flood events considered. Values of average velocity and shear stress were plotted versus Mannings n to identify the critical river cross-sections that will control the rock size design and to identify any trends associated with the increase or decrease in roughness. The results indicated that the highest velocity and shear stress values occur between the two cross-sections where the flood plain narrows and the channel slope is the steepest. Shear stresses in this segment were more than twice those at other locations. Also, the shear stress at this location increased rapidly as the Mannings n was decreased especially for the higher discharges. A comparison of the median rock diameters required at Slickrock with those at Grand Junction showed that rock sizes at Slickrock site were roughly twice those at Grand Junction even though the flood discharges were much less. This was primarily a result of the steeper energy slope at Slickrock.

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.

Subtask C. Riprap Protection For Local Drainage Channels

No work was performed on this subtask during the quarter. The literature review has been completed. Current plans are to publish both the subtask B and C results in the same report.

Subtask D. Vegetation Covers

The final report for this subtask has been completed.

Subtask E. Economics of Riprap Design Procedures

The final report for this subtask was completed and forwarded to the NRC this quarter.

Sbtask F. Watershed Model Evaluation

Work on this subtask continued through the quarter. The computer test runs for the overland erosion model, CREAMS, were completed. Rainfall-runoff and erosion data for two watersheds in Arizona were obtained from the Agricultural Research Service in Tuscon, Arizona. One of the data sets will be used to evaluate the use of overland erosion models at uranium tailings sites.

Subtask G. Rock Selection and Durability

The report will be completed next quarter. The initial draft report for this subtask was completed and underwent peer review.

Subtask H. Impoundment Slope Stability and Settlement

The initial draft of the subtask report has been completed and is currently undergoing review and rewriting.

PROJECTED WORK FOR NEXT QUARTER

The reports for subtasks A, B and C (combined), and G should be completed prepared for clearance and final review. Work will continue on the revisions of the subtask H draft report and on the engineering handbook.

JOURNAL ARTICLES

A refereed journal article by W. H Walters, "Computational Methods for Estimating Overland Erosion," was prepared and submitted to Oak Ridge National Laboratory for publication in the journal Nuclear Safety later this year.

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

Evaluation of the Wyoming field tests was completed with the final monitoring trip during the first week of June. Final evaluations of both the chemical stabilizer test plots and the windscreen test panels were made at that time. Windscreen materials were left in place in case another later trip to the site is feasible.

The PNL windscreen field test is proceeding without problems. Monitoring is expected to be complete by the end of July. Data are showing marked differences in the effectiveness of windscreens to reduce wind velocity across a simulated tailings pile. Results of these field tests will be presented in the final report, which is being prepared.

PROJECTED WORK FOR NEXT QUARTER

Evaluation of the PNL field test will be completed. The results of the field tests will be presented in the final report, which should be completed during the next quarter.

REPORTS ISSUED DURING THE QUARTER

A journal article on this project has been prepared and accepted for publication in Nuclear Safety. It is to appear in the Nov.-Dec. issue. Also, a paper has been accepted for presentation to a co-sponsored (EPA-EPRI) symposium on particulate control technology to be held in August in Kansas City, Missouri.

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 evaluate approaches to dewatering tailings pits to reduce potential seepage into ground water and to enable early long-term placement of thick earthen covers over the hydraulically filled tailings pits. Special attention is given to evaluating linear consolidation analysis as a worst-case estimator of settlement during tailings dewatering. Measurement techniques for establishing void ratio and degree of saturation as functions of the stress-state parameters will be evaluated and tested. Hydraulic conductivity as a function of void ratio will also be evaluated.

ACCOMPLISHMENTS DURING PAST QUARTER

Measurements of partially saturated consolidation are continuing. An experiment with slimes material and a load stress of 0.2 m has been completed. A second slimes sample is presently being tested with a load stress of 6.0 m and is nearly done. The next slimes sample will be loaded to a stress of 25.0 m.

We have decided to give a name to the modified version of TRUST that has been used in this task. The new name is TRUNC, which stands for TRANSient, UNSaturated Consolidation. In April a presentation was given to Frank Swanberg, Georgio Gnugnoli, and Bill Dam which illustrated some comparisons between TRUNC predictions and simpler, empirical calculations. For the consolidation of slimes, the empirical calculations predicted settlements about equal to TRUNC settlement predictions. Our findings also indicated that TRUNC may be the only means of arriving at estimates of the time of partially saturated settlement because no empirical solution exists at the present time. If the magnitude of partially saturated settlement proves to be significant, then TRUNC is the only means currently available to estimate time of cover placement.

The empirical calculations and computer analysis have been completed for the report due in September. A draft of the document, including the appendices, has been through a peer review and is now being edited.

PROJECTED WORK FOR NEXT QUARTER

The partially saturated consolidation tests will continue. Work has begun and will continue on one- and two-dimensional studies of the interactions between consolidation and fluid flow. Two materials will be considered,

on settlement. The results of these studies will form the basis for one of the December reports. Both December reports will be drafted during this quarter.

CHANGE IN DELIVERABLES

In talks with Swanberg and Gnugnoli, we have decided to change the nature of the remaining deliverables.

Original Deliverables:

1. "TRUST-II: A Computer Program for Variably Saturated Flow in Deformable Media" July, 1984. Discussions with T. N. Narasimhan have indicated that the use of the name "TRUST-II" would not be acceptable. Because the major part of the code being used in Task 3 has already been documented as TRUST, we decided to publish the changes we have made to the code in an appendix to a new report due in September, 1984. The modified code will be called TRUNC.
2. "The Sampling and Measurement of Uranium Mill Tailings Consolidation Characteristics", September 1984. As mentioned in previous reports, the measurement of partially saturated consolidation has taken longer than anticipated. In order to give us a little more time to generate sufficient data, we have decided to delay the publishing of the data until December.
3. "The Analysis of Drainage and Consolidation at Typical Uranium Mill Tailings Sites", December 1984.

New Deliverables:

1. "Consolidation of Uranium Mill Tailings: A Comparison of Analysis Techniques" (tentative title) September, 1984. The objective of this report will be to compare the predictions of settlement and settlement times between TRUNC and an empirical approach. This analysis will be done for both the saturated and partially saturated cases. This report was conceived to answer the questions raised by Swanberg and Gnugnoli as to which analysis techniques were simplest yet appropriate for different conditions. This report will contain an Appendix outlining the modifications made to TRUST. A new input manual will be included, as well as a section with valuable suggestions for solving problems that arise when using the modified TRUST code.
2. "The Sampling and Measurement of Uranium Mill Tailings Consolidation Characteristics", December 1984.
3. "The Analysis of Drainage and Consolidation at Typical Uranium Mill Tailings Sites", December 1984.

100% slimes and a mix of 60% sands and 40% slimes. In addition, several two-dimensional simulations will be conducted to gauge the effect of drain spacing on settlement. The results of these studies will form the basis for one of the December reports. Both December reports will be drafted during this quarter.

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2. "The Sampling and Measurement of Uranium Mill Tailings Consolidation Characteristics", December 1984.
3. "The Analysis of Drainage and Consolidation at Typical Uranium Mill Tailings Sites", December 1984.

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

All ongoing laboratory experiments were completed during this quarter. Results of the contaminant/carbonate complexation studies and the tailings radium attenuation experiments are being compiled and will be documented in a laboratory progress report. Following the laboratory progress report, a final report covering the studies completed over the duration of the Tailings Neutralization task will be submitted. At this time, no further laboratory experiments are expected to be initiated before completing the final report for Task 4.

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 FY83. The plan outlines three alternatives for field testing the most promising and cost-effective current neutralization technique.

REPORTS ISSUED DURING THE QUARTER

None.

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

Task Project Manager: R. W. Nelson
Principal Investigators: T. J. McKeon
C. J. Hostetler

FIN 8-2370 (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 leaching by the ground water. The second method currently being studied is disposal of tailings in deep underground mine stopes below the water table.

ACCOMPLISHMENTS DURING PAST QUARTER

The analysis method developed during the first quarter of this year for evaluating the environmental consequences resulting from the disposal of uranium tailings in deep underground mine stopes was extended. The extension enabled determination of overall downgradient leachate plume sizes and shapes. The results obtained represent two levels of detail. The first overview evaluation provides the maximum plume width, maximum depth of plume, and an easily used multiplier index of the plume length. These results are provided as a set of six summary graphs for direct use. The second or more detailed evaluation of plume configuration evaluation involves an interactive computer code, which provides the detailed downgradient plume shape. The leachate plume concentrations that result when no chemical interaction with the aquifer material occurs are also provided by the interactive computer code.

The interactive code was used in slightly modified form to provide a summary of maximum plume width and depth as well as the plume length index. The overall plume dimensions are summarized in dimensionless graphical form and demonstrate those dimensionless parameters that affect the leachate plume size. The plume size and shape are affected most by the contrast ratio between the tailing's and aquifer material's hydraulic conductivities followed closely by the orientation of the mine stope with respect to the regional ground-water gradient. Though the plume width mostly depends on the stope length-to-diameter ratio, both the maximum plume depth and length index are only mildly dependent on this ratio. The leachate plume dimensions were also found to be only slightly dependent upon the regional ground water gradient, U . In the range of $0.0005 \leq U \leq 0.005$ commonly encountered, the plume depth and length are not affected by the gradient, and as long as the stope length to diameter ratio is less than 100. the plume width changes by less than 5%.

The graphical summary results are used to locate and to help determine the highest expected concentration in the downgradient water supply well used in the consequence analysis. The graphical results show the maximum depth of leachate plume and allow the initial length of the well perforation in the water supply well to be selected so as to intercept the most tailings leachate from the plume downgradient of the stope. Using the maximum plume depth, the well pumping ratio per unit length of perforated well depth is calculated; from these calculations, the well capture width is obtained directly. If the well capture width is less than the leachate plume width, a satisfactory consequence evaluation for the contaminant concentration in the water pumped from the well will be obtained. However, if the capture width of the well exceeds the plume width, dilution will occur in the pumped well. In this case, through using the graphical results for leachate plume width, the well pumping rate can be reduced as needed until the capture width for the well just equals or exceeds the plume width. In this way a consequence evaluation yielding the highest expected concentrations in the pumped well water can be determined.

The fluid flow aspects discussed and the methods extended for determining the leachate concentrations leaving the tailings-filled stope are essentially completed. The report outline for Leachate Plume Migration Downgradient of Uranium Tailings Disposal in Deep Mine Stopes, describing this work is completed and writing of the report will be accomplished during the next quarter. There remains the very important geochemical and coupled transport code evaluation using the column data from companion experimental tasks. Then methods will be provided for incorporating those predictive results into the leachate flow results already completed.

The modifications of the initially coupled transport and geochemical code have been completed. This involved providing updating capability for the ionic strength at each time step, providing ways of differentiating between precipitates that were sorption surfaces and those already present in the media, and extending the ion-exchange capability of the code. Task 5 assisted in the funding of these modifications, which were accomplished at a slower rate than originally expected. Also, several numerical problems arose as a result of the different rates of advance of particular chemical species and had to be overcome. Accordingly, the expected testing of the code using the experimental column data still remains to be done during the next quarter.

PROJECTED WORK FOR NEXT QUARTER AND THE SUBSEQUENT QUARTER

The report, Leachate Plume Migration Downgradient of Uranium Tailings Disposal in Deep Mine Stopes, will be prepared and submitted for clearance. This report will present all the fluid flow analysis results and methods developed to the present time. This includes the methodology for evaluating the leachate concentrations as the ground water emerges from the tailings.

The initial testing of the extended geochemical/transport code using the experimental column data will be carried out and completed during the next quarter. Specifically, the code will be used to calculate the expected

effluent concentrations when a constant tailings leachate feed solution interacts with the aquifer materials. The calculated effluent concentrations will then be compared with the laboratory measured outflow concentrations.

If the calculated and measured effluent concentrations show appropriate agreement, two additional tasks are required. First, the extended geochemical/transport code will be used to calculate the chemical interactions, precipitation, sorption, and finally the long-term continuing leachate concentrations in the migrating ground water that will occur under the conditions of the time-varying leachate concentration. Specifically, the experimental columns had a constant leachate feed solution concentration, whereas in the actual field, the leachate concentrations leaving the tailings begin at a high initial concentration but gradually diminish over time as more ground water continues to leach the tailings. The geochemical/transport model will be used to evaluate the effects of the diminishing leachate concentration moving through a long column of aquifer material. The second step involves incorporating these leachate aquifer results into the evaluation methods to provide the overall environmental consequences.

REPORTS ISSUED DURING THE QUARTER

None.

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 whether site and structure decontamination are 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).

ACCOMPLISHMENTS DURING PAST QUARTER

This task was not funded in FY84, and exhausted its funds carried over from FY83 in December. No work was performed in the last quarter except for preparation of final reports.

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.

- 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.
- Prepared a presentation to be made to the National Research Council Panel on Uranium Mill Tailings to be held in July.

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, January 1984 to March 1984. PNL-5015-2, 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 an NRC regulatory guide.

ACCOMPLISHMENTS DURING PAST QUARTER

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

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

PROJECTED WORK FOR NEXT QUARTER

This project was scheduled for completion in CY83. 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 are obtained.

REPORTS ISSUED DURING THE QUARTER

None.

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

This task exhausted its uranium recovery funds carried over from FY83 in February, 1984 and is now funded by Low-Level Waste.

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
L. E. Eary
S. B. McLaurine
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

This project was reviewed by NRC in Silver Spring, Maryland on April 24, 1984. The history, accomplishments, and future plans of the project were presented to Uranium Recovery Field Office personnel in Denver, Colorado, on April 23, 1984.

A no-cost extension of the period of performance of this project to an estimated date of December 31, 1984 has been approved by NRC. This will enable us to complete the laboratory work this summer and write the final report by the end of the calendar year.

Task 1. Site Characterization

No new work was performed for this task during the past quarter. The only additional planned work for this task is to characterize the clay minerals present in the sediments used in the batch and column experiments. The characterization of the clays will include a determination of their cation exchange capacity and an analysis of the exchangeable cations present on the clays under aquifer conditions. The importance of these measurements will be discussed in the following section.

Task 3. Natural Restoration Processes

The results of the natural restoration experiment using Wyoming sediment and synthetic lixiviant are being used to develop a computer chemical model

of aquifer restoration in response to natural chemical processes. The natural restoration experiment was described in the last quarterly report. To summarize the results of the experiment, we found that the initial effluent samples from the columns had much higher concentrations of calcium, sulfate, and magnesium, and a lower pH than the original ground water in the aquifer. As the synthetic lixiviant displaced the residual pore water in the column, the calcium, magnesium, sulfate, and uranium concentrations decreased and the sodium concentration and pH increased. However, influent concentrations of dissolved constituents are not attained until many pore volumes of solution have passed through the column, suggesting that chemical processes are important in understanding the migration of the dissolved load. As Figure 1 shows, the sodium value did not reach its influent concentration of 414 ppm until over seven pore volumes of solution flowed through the column. The calcium and magnesium concentrations show a corresponding decrease to influent values of 79 and 44 ppm, respectively. This effect was probably caused by ion exchange between the solution and clays present in the sediment. To properly model this system, we must know the cation exchange capacity of the clays, and we must know composition of ions initially present on the clays at the beginning of the experiment. In the last quarterly report we pointed out that the uranium concentration increases slowly during flow and approaches influent concentration by the eighth pore volume. Because the dominant uranium species in this system is probably an anionic carbonate complex, it is doubtful that ion exchange with clays is affecting its behavior. However various oxyhydroxide solids are present that could adsorb the uranium carbonate complex, thereby retarding its movement through the column.

From the solution data, we are developing conceptual chemical models of the system that will incorporate the reactants and chemical processes that appear to be important. The models will include such processes as 1) oxidation of pyrite to produce ferric oxyhydroxides, sulfate, and hydrogen ion; 2) dissolution of magnesian calcite; 3) precipitation of gypsum; 4) equilibration of the solution with a gas phase having a fixed carbon dioxide gas partial pressure; 5) ion exchange involving calcium, magnesium, and sodium; 6) adsorption/desorption of uranium onto ferric oxyhydroxide; and 7) aqueous complexation of dissolved components. MINTEQ is being used to make the necessary calculations. The results will be compared to the available analyses of solution effluent samples to determine the adequacy of the model for simulating the chemical processes occurring during storage of the sediment and as natural restoration takes place.

The batch experiments with Texas and Wyoming sediments that began in February, 1984 were completed in mid June. In these experiments we evaluated the effect on solution composition of long-term (up to 3 1/2 months) storage of lixiviant and sediment. Some of the sediments were amended with pyrite and FeS-coated sand to compare with the naturally occurring pyrite in the sediment. The solution composition data are being compiled at this time, and the results will be discussed in the next quarterly report.

FIGURE 1
EFFLUENT CONCENTRATIONS OF CA, MG, AND NA FROM COLUMN 2
OF THE WYOMING NATURAL RESTORATION EXPERIMENT

Task 4. Induced-Restoration Techniques

During the past quarter, a series of batch experiments were conducted to test the effectiveness of sodium sulfide as a reducing agent added to spent lixiviant. The purpose of adding a reducing agent to the solution is to re-move dissolved oxygen from the spent lixiviant, which would then be recirculated through the leached ore during aquifer restoration. If dissolved oxygen is present in the recirculating solution it might mobilize trace metal contaminants (uranium, arsenic, selenium, and molybdenum) present in the sediment. Sodium sulfide was added to various solution types and solution/solid mixtures that consisted of distilled, deaerated water, distilled water, lixiviant, lixiviant with iron-oxide-coated sand, and lixiviant with leached ore. The initial sulfide concentrations for each set of experiments were 0.01, 0.001 and 0.0001 molar (330, 33, and 3.3 ppm as HS, respectively). The pH and Eh of the solutions were monitored and solution samples were taken periodically to measure changes in sulfide, oxygen, and uranium concentrations.

The addition of sodium sulfide raised the pH and lowered the Eh of all the solutions. The amount of change varied with the type of solution and the amount of sodium sulfide added. In general, the distilled, deaerated water systems showed the biggest change [+5 pH units and -700 MV Eh (final Eh = -300 MV)] because they are poorly buffered and less well poised. The systems with lixiviant and lixiviant with sediment showed a less drastic pH/Eh change. However, in the 0.01 molar sulfide lixiviant/sediment case, the pH still increased by 3 units, whereas in the 0.001 molar sulfide case for lixiviant with iron-oxide-coated sand, the pH change was more modest, 0.5 pH units. In all systems, with 0.01 molar sodium sulfide the Eh dropped from +300 MV to approximately -300 MV when the reducing agent was added. In all cases, except the lixiviant with leached ore, the Eh dropped to -150 MV when 0.001 molar sodium sulfide concentrations were used. For lixiviant with leached ore the initial Eh drop was to approximately +200 MV. The oxidized leached ore apparently contained constituents that oxidized sulfide, thereby poisoning the Eh at a higher value than possible with the other systems.

The objective of adding a chemical reductant to the system is to remove oxygen from solution so that it will not mobilize trace metals found in the sediment. The low Ehs measured imply that sulfide removed oxygen from solution and accomplished this goal. We measured dissolved oxygen concentrations of the solutions at the end of each experiment using the Winkler titration method. In almost all cases, the addition of sulfide to the solutions reduced the oxygen concentration from 10 ppm to less than 0.1 ppm. In a few cases, we found measurable oxygen but this may have been introduced during filtering of the sample prior to titration. Another indicator that the chemical reductant is having the desired result is changes in dissolved uranium concentration after sulfide addition. The largest changes observed for uranium concentration were found when the most sulfide was added. For the 0.01 molar sulfide systems, one day after sulfide addition the uranium concentration had dropped from 42 mg/L to 1 to 10 mg/L depending on the type of solid present. After 48 hours the uranium concentration dropped further to 0.1 and to less than 0.02 mg/L for some cases. Interestingly, the uranium concentration increased to greater

than 1 ppm a few days after reaching its minimum values. This may have resulted from atmospheric oxygen contamination or perhaps because of slow oxidation reactions occurring between the sediment and solution.

From the initial results of the batch experiments on chemical reductions, it appears that sodium sulfide does many of the things that would enhance aquifer restoration. However, it is also important that the addition of sodium sulfide does not produce adverse side reactions. In this case, the increase in pH may prove to be a problem. Carbonate minerals are less soluble at high pH and their precipitation in the system could potentially lower the permeability of the aquifer, thereby inhibiting the movement of restoration fluids. In the case in which 0.01 molar sodium sulfide was added to lixiviant, the pH is raised from approximately 8 to 10.5. The dissolved calcium concentration decreased by 250 mg/L suggesting that calcium carbonate is precipitating. When smaller amounts of sodium sulfide are added to solution, the pH change is much less drastic and there is a correspondingly lower change in dissolved calcium values. Another possible adverse side effect to adding sodium sulfide could be the swelling of clays in the sediment because of increased dissolved-sodium concentration. This could also reduce the permeability of the aquifer. However, most of the natural ground waters found in aquifers used for in situ leach mining are dominated by sodium; therefore, the addition of reasonable amounts of sodium to solution should not cause an appreciable problem. Additional experiments using a flow-through column apparatus are being planned to further evaluate the applicability of using sodium sulfide to enhance aquifer restoration.

During the past quarter, the Twin Cities Research Center of the Bureau of Mines completed a mine operator survey of experiences with surface treatment restoration equipment. They contacted 17 companies that have 27 in situ leach operations. Surface treatment methods had been used at 14 sites. Reverse osmosis had been used at 9 sites; electrodialysis at 3 sites; and ion exchange at 2 sites. Some of the general comments made concerning these various methods of solution treatment were

Reverse Osmosis - generally effective and dependable; some units subject to fouling; somewhat limited pH range.

Electrodialysis - very effective; reliable; high initial capital cost.

Ion Exchange - effective and economical for selective contaminants

PROJECTED WORK FOR NEXT QUARTER

Column experiments will be conducted with leached ore and lixiviant to which sodium sulfide has been added to more closely simulate actual restoration by recirculation aided by a chemical reductant. The pH and Eh of the effluents will be monitored and solution analyses will be made for major, minor, and trace constituents. The results of the long-term (3 1/2 month)

batch experiments with Texas and Wyoming sediments will be analyzed and compared with previous similar experiments. The MINTEQ-model of natural restoration will be further developed and compared with solution analyses for correctness.

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None.

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