Interim Report

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

**Quarterly Progress Report
January - March, 1982**

April 1982

**Prepared for
the U.S. Nuclear Regulatory Commission
under Contract DE-AC06-76RLO 1830**

**Pacific Northwest Laboratory
Operated for the U.S. Department of Energy
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Interim Report

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

QUARTERLY PROGRESS REPORT
JANUARY - MARCH, 1982

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April, 1982

Prepared for the
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under Contract DE-AC06-76RL0-1830

Pacific Northwest Laboratory
Richland, Washington 99352

EXECUTIVE SUMMARY

Progress is reported in ten areas concerned with characterizing, controlling, and measuring airborne emissions and liquid waste from uranium recovery mills and in situ mining.

Work progressed on developing the data base for a riprap engineering handbook for long-term stabilization of tailings pile radon suppression covers. A report on environmental considerations was completed and a report on selection and durability of rocks neared completion. A report on rock riprap design methods and their applicability to long-term protection of uranium mill tailings was in the review stage, and a fourth report will soon be completed on wind erosion and deposition considerations. The information in these reports will provide important inputs to the handbook for engineering riprap protection of tailings piles.

The interim stabilization study was directed toward obtaining additional information from mill owners who had tried dust suppressing methods and materials. Additional commercial products were obtained and laboratory and field study plans developed to screen and test the more promising materials.

Emphasis was placed on overcoming the limitations of classic models of consolidation during tailings dewatering through identifying simplifying assumptions and considering ways that parameters of large strains and their non-linearity, anisotropy of tailings, and other soil physics considerations could be realistically accounted for in models. Approaches of other researchers were reviewed.

A report of a study of materials and installation procedures was issued as NUREG/CR-2608, PNL-4150. The report showed how accepted drainage practices for agriculture might be applied to tailings impoundments. Differences in the systems will significantly limit a direct application of agriculture drainage designs to tailings dewatering.

The relevance of acid mine drainage neutralization methods to uranium tailings was explored and similarities were identified. Potential for

segregation and encapsulation with asphalt or cement was studied and possible use for isolating the more radioactive constituents considered. Literature study of precipitation agents continued.

The study addressing the ramifications of below ground water disposal made considerable progress in obtaining water retention, particle size and several chemical characteristics of tailings and soils obtained from three mill sites.

The variably saturated flow code TRUST was used to investigate three disposal pit scenarios; all pits were assumed to have a common geometry in sandstone, backfilled with 30 feet of overburden, then covered with three feet of clay with a stub wall rising to 20 feet along the pit sidewall. Four gravel-enclosed drains were assumed to be placed above the clay liner. The three scenarios differed primarily in the depth to the regional water table and in the second scenario a regional water gradient of 0.005 was assumed. Of considerable interest to the objective of this task was the result that for scenario three in which the regional water table was 74 feet above the bottom of the pit, about 2.5 times more water enters the drains near the stub wall clay liners. This result suggests that the configuration for scenario three may permit better overall control of the tailings slurry water. Other hydrologic effects were obtained.

The retardation from the buffering capacity of the clay was dramatically shown with the modeling effort; the clay liner effectively caused precipitation of essentially all of the contaminants.

Radon and radon daughter measurement instruments and techniques were under evaluation and testing during the reporting period. Planning and instrument selection for the indoor-outdoor measurements were the principal activities for this subtask. Track etch detector performance was evaluated with significant effort expended in cross calibration and improvements in their use.

Literature review was completed of concentration limits and measurement methodologies for uranium, ^{226}Ra , ^{230}Th , and ^{210}Pb .

Methods and instrumentation for well-logging were reviewed and a conclusion reached that well-logging had been applied, if at all, to a very limited extent

for remedial action programs. A uranium tailings pile simulator for laboratory sensitivity tests was readied and arrangements made with a mill operator to case two holes through their tailings pile for well logging demonstrations.

Experience with contamination from uranium mill tailings and the variability of soil concentrations in studies to date were investigated in a literature review. The study of measurement technologies for surface and ground water constituents was initiated and information gathered on current practices and the statistical aspects of sampling.

In the leachate movement investigation characterization of additional tailing solutions was completed and state-of-the-art methods were developed and applied to the analysis of these solutions. A geochemical model for use on the chemical modeling portion of the study was selected and verification begun.

A substantial part of the findings to date on radon diffusion and attenuation in soil was consolidated in three topical reports: Radon Attenuation Properties of Candidate Cover Materials, Comparison of Radon Diffusion Coefficients Measured in the Field to Laboratory Measured Values, and A Mathematical Model for Radon Diffusion in Earthen Materials. Issue dates are expected to be June, July, and May, 1982, respectively.

Laboratory measurements were performed to show the effect of tortuosity on the diffusion coefficient and to confirm the numerical value of this coefficient.

Three candidate cover soils for possible use at the Grand Junction tailings pile were obtained and are being evaluated for radon attenuation in the laboratory attenuation test facility to determine their effectiveness for radon suppression.

An important accomplishment in the study of contamination control in in situ recovery of uranium was the acquisition of several kinds of samples from Uranium Resources Incorporated in situ operation in south Texas. A cooperative agreement with Resources Incorporated provided PNL with access to the operation. Many measurements were taken at the site on carefully sampled lixiviant and ground water. Well cores were obtained in such a way to protect the cores from unduly oxidizing.

During the reporting period monthly administrative reports were submitted to the NRC-RES. A program review was conducted at PNL, Richland, on March 2 and 3 and reviews scheduled for the second quarter at NRC Headquarters.

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INTRODUCTION

Pacific Northwest Laboratory (PNL) has in progress several discrete investigations for the U.S. Nuclear Regulatory Commission (NRC) concerned with process wastes during the active life of uranium mills and during an extended period following their shutdown. 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 in using rock covers (riprap) as well as their design for armoring tailings pile covers, shorter term stabilization options for controlling windblown particles, leachate movement in soil, tailings dewatering, disposal deliberately below the water table, neutralization incentives, contamination control and restoration in in situ uranium recovery and effluent and environmental measurements, instrumentation and protocols. It is expected that many results of these studies will be used in developing regulatory guides and better evaluation of environmental impacts during and following the active life of a uranium recovery facility.

This is the third progress report in the current format. The first report (September to October, 1981) carries the basic report number PNL-4105. This and succeeding reports will be identified with a dash number -1, -2, etc., corresponding to the quarter commencing with Fiscal Year 1982. Progress reports covering periods prior to July, 1981 are letter reports addressed to the sponsor.

Progress on each of several separately identified areas is reported in the following sections. The information reported is preliminary and conclusions should be regarded as tentative. About 20 topical reports are scheduled during the year to provide detailed description of phases of the work and conclusions therefrom. The authors solicit comment and suggestions from readers of progress and topical reports.

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

Project Manager: L. C. Schwendiman

FIN B-2370

Objective

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

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TASK 1. LONG-TERM STABILIZATION

Task Project Manager: J. Mishima

Principal Investigator: J. Mishima

Support Investigators: J.K. Young (Site Characteristics)
L.W. Long (Site Characteristics, Rock Durability)
T.J. Bander (Wind Erosion, Soil Deposition)
W.H. Walters (Water Erosion, Riprap Engineering)
G.B. Parker (Riprap and State-of-the-Art)
G.C. Lindsey (Rock Durability, Archaeology)
B.K. Marshall (Editor)

FIN B-2370 (Task 1)

Objective

Acquire, evaluate, and assess information on the specification and proper installation of riprap ("rock armor") and other materials for long-term protection of stabilized mill tailings from water and wind erosion, under a full range of site and environmental conditions.

Accomplishment During Report Period

Subtask A. Baseline Data

1. Site Characteristics. The document, "Environmental Factors Affecting Long-Term Stabilization of Radon Suppression Covers of Uranium Mill Tailings," has been submitted to NRC publications for issuing. Some of the pertinent findings are shown in Table 1.
2. Wind Erosion and Soil Deposition Models. Chepil's Wind Erosion Equation developed for estimating losses from agricultural plots appears to be the most applicable for losses from covered uranium mill tailings. The equation considers the principal elements affecting wind erosion and has the form:

$$E = f(I, K, C, L, V)$$

where:

E = soil loss by wind, tons/acre/year

I = soil wind erodibility

K = roughness factor.

TABLE 1. Disruptive Parameters Summarized by Region

	<u>EASTERN WASHINGTON</u>	<u>WYOMING BASINS</u>	<u>COLORADO PLATEAU</u>	<u>TEXAS GULF COAST</u>	<u>SOUTHERN ROCKIES</u>
NO. OF PONDS					
ABOVE-GROUND	2	9	10	NA	2
BELOW-GROUND	1	4	4	NA	1
RUNOFF POTENTIAL	LOW-HIGH	MODERATELY LOW-HIGH	MEDIUM TO HIGH	LOW	HIGH
PROBABLE MAX. PRECIPITATION	*	56 cm IN 6 HR	60 cm IN 6 HR	*	33 cm IN 1 HR
WIND EROSION POTENTIAL	HIGH	MEDIUM	MEDIUM TO HIGH	MEDIUM	MEDIUM
MAXIMUM GUST (m/sec)	32	33-36	20-36	32	31
TORNADO POTENTIAL	LOW	LOW	LOW	MODERATE	LOW
MAX. EARTHQUAKE ACCELERATION (g)	0.1	0.03-0.05	0.05-0.4	0.06	0.2

*NOT AVAILABLE

TABLE 1. Disruptive Parameters Summarized by Region (contd)
(Definition of Parameters)

NUMBER OF PONDS -

RUN-OFF POTENTIAL - THIS IS A QUALITATIVE TERM TAKEN FROM LITERATURE WHICH IS A JUDGMENT BASED ON SOIL PERMEABILITY, SLOPE, PRECIPITATION, AND GROUND COVER

WIND EROSION POTENTIAL - THIS IS A QUALITATIVE TERM TAKEN FROM LITERATURE BASED ON SOIL TEXTURE; LOW RANGES FROM NONCALCAREOUS CLAY LOAM TO SILTY CLAY, AND HIGH RANGES FROM SANDY LOAM TO SAND

MAXIMUM GUST - RANGE OF VALUES FOR SITES FROM LITERATURE WITH A 100 YR RECURRENCE PROBABILITY

PROBABLE MAXIMUM - THE PHYSICAL UPPER LIMIT OF STORM RAINFALL FOR THE SITE AREA BASED ON
PRECIPITATION STATISTICAL ANALYSIS WHICH WHEN CONVERTED TO FLOOD FLOW, RESULTS IN MAXIMUM
FLOOD CONDITIONS

TORNADO POTENTIAL - AN ESTIMATE BASED ON MEAN ANNUAL FREQUENCIES OF TORNADOES FOR A 1° SQUARE AREA. LOW RANGES FROM 0-1, MODERATE RANGES FROM 1-2, AND HIGH RANGES FROM 2-5

MAXIMUM EARTHQUAKE - MEASUREMENT OF STRESS INDUCED IN THE IMPOUNDMENT STRUCTURE MEASURED IN G's
ACCELERATION DUE TO GROUND SHAKING FROM THE MAXIMUM EARTHQUAKE EVENT

C = local wind erosion climatic factor
 L = equivalent field length
 V = vegetation factor.

Table 2 taken from Smith, Whicker, and Myer (1982)^(a) indicates how the primary environmental variable determined by agricultural scientists to be important for wind erosion are incorporated into the Wind Erosion Equation. Many other models are available but are not as applicable (Smith, Whicker and Meyer 1982, Table 2).

TABLE 2. Environmental Variables and Model Parameters for the Wind Erosion Equation*

<u>Primary Wind Erosion Variables</u>	<u>Equivalent Erosion Equation Parameters</u>
Soil Erodibility Index, I Knoll Erodibility, I_s	Soil and Knoll Erodibility, I
Surface Crust Stability, F_s	Disregarded - Crust is Transient
Soil Ridge Roughness, K_r	Soil Ridge Roughness Factor, K
Average Wind Velocity, v Surface Soil Moisture, M	Local Wind Erosion Climatic Factor, C
Distance Across Field, D_f Sheltered Distance	Field Length, L
Quantity of Vegetative Cover, R Kind of Vegetative Cover, S Orientation of Vegetative Cover, K_o	Equivalent Vegetative Cover, V

*Smith, Whicker and Meyer (1982)

(a) W. J. Smith, F. W. Whicker, and H. R. Meyer. 1982. "A Review and Categorization of Particulate Entrainment Models."

Two general types of models for soil deposition (source depletion and surface depletion models) have been found in the literature. Neither type appears adequate for estimating long term deposition of soil on riprapped surface since both are for low particulate concentrations and do not consider the range of environmental variables (surface roughness, slope, fluid flow) that are important.

3. Overland Flow Erosion. Data accumulation is in progress on models to describe erosion from rain impact and runoff.
4. Channel Flow and Riprap Design. Initial work commenced on the development of the basic design procedures for the riprap blanket. A design procedure was outlined in preparation for some initial case studies using two (or more if necessary) mill tailings sites to test the proposed methodology on paper for completeness. The primary purpose is to identify possible gaps and problem areas in the procedure. The basic design procedure is outlined below:

PROPOSED DESIGN PROCEDURE

1. Determine Erosion Potential at the Site
 - Determine Overland Erosional Stresses and Erosion Rate
 - Probable Maximum Flood (PMF) Study
 - Peak Flow Discharge
 - Perform Backwater Analysis (if PMF a Design Criteria)
 - Design Velocity
 - Water Surface Elevation
 - Decide on Protection Requirements Based on Site Hydrology and Erosional Characteristics
2. Field Reconnaissance
 - Select Rock Type and Source
 - Based on Guidelines
 - Laboratory Tests
 - Availability

- Determine Potential Size Reduction Due to the Weathering Process
 - to be Considered in Stone Size Determination
3. Riprap Design
- Determine Average Rock Size (PMF Criteria)
 - Use Selected Design Procedure
 - Incorporate Potential Size Reduction
 - Determine Stone Graduation from Guidelines
 - Design a Riprap Layer
 - Based on Guidelines and Impoundment Design
 - Results from Flume Studies to be Incorporated into Guidelines (Non-Uniform Flow Problems)
 - Determine Size of Material to Resist Overland Erosion (if PMF is not a Design Criteria)
4. Final Design of Cap and Riprap
5. Construction Specifications
5. Rock Durability and Selection. The document, "Long Term Survivability of Riprap for Armoring Uranium Mill Tailings and Covers: A Literature Review," is in clearance and should be issued by mid-April. The significant findings are:
- rock durability must be viewed in terms of the time required for its survivability and the criteria it must meet for its intended application: for our purposes the rock must survive for up to 1,000 years or greater and it must be able to maintain a sufficient size (weight) to resist incipient motion in the event of occurrence of the probable maximum flood (PMF).
 - ample evidence exists of the inability of many rock types to survive less stringent criteria than those required for mill tailings armoring
 - all rocks weather, though at different rates and to different extents depending upon the environmental conditions in which the rock is placed and the characteristics of the rocks themselves; climate and lithology control the weathering processes

- physical and chemical weathering agents are active in processes of rock deterioration; in general, the regions of the U.S. where most uranium mills are found are subject to physical weathering (e.g. frost action, salt crystallization, temperature induced volume changes, etc.); weight loss can be severe and rapid under the right conditions
- tests for durability do not address the question of time, because most applications of commercial grade stone assume a) relatively short project lives, and b) periodic maintenance; actual rock performance as a function of time is an area not adequately addressed in the literature
- quarrying practices may play as important a role as any other factor in insuring rock durability (particularly for riprap sources)
- many durable rock types exist (e.g. dense igneous materials) which should be suitable for mill tailings armoring. Less durable varieties (e.g. most sedimentary varieties) may prove to be more plentiful and thus require additional investigation.

Our efforts on the literature review have been supported by Dr. Richard Jahns of Stanford University who has prepared his own report to us which appears as an appendix to our document. His findings and conclusions support our own.

6. Survivability of Ancient Manmade Earthen Mounds. Data accumulation is essentially completed and preparation of the manuscript is in progress. The following observations can be made based upon the information at hand.

- Mounds and other structures are found in many parts of the world and are the product of a variety of cultures; they usually take the form of ceremonial structures (e.g. burial sites or effigy mounds) or serve as large platforms upon which buildings are placed.
- the survivability of mounds appears to contradict currently accepted theories of natural processes. Large horizontal effigy mounds built hundreds of years ago standing only one meter high should have eroded to the point of being indistinguishable from the surrounding countryside, but such is not the case

- the mound builders have employed a variety of design and construction techniques and the evidence indicates that they learned from their mistakes, i.e. their ability to erect a structure which could survive natural erosional forces got better with time (a "learning curve") and practice
- vegetation was not deliberately introduced; it occurred after the site was abandoned
- in the southwestern U.S. and other parts of the world characterized by arid or semi-arid conditions, the construction of ceremonial, agricultural, and dwelling sites relied upon the use of stone rather than soil, sandstone was a popular building material in the Four Corners region and the Colorado plateau area.

Subtask C. Studies in Support of Guide Development

Efforts to develop study plans in two areas (physical models studies in flumes and rock selection procedures) continues. The tentative plan for the rock selection procedures studies are:

- identification of rock types available on a commercial basis in uranium mill regions
- identification of available data in the historical application and performance of these rock types
- selection of representative rock types for further study
- analysis of performance as a function of time
- investigation of performance with available testing procedures
- simulation of performance with available weathering and durability tests.
- correlation and verification of predicted vs. actual performance.

The number of rock types and subtypes we will be able to study depends upon a) availability of data, and b) time and cost constraints on performing necessary tests and data analysis. In general, we plan to focus upon those rock types most readily available and which we believe a priori may pose durability

problems. Sandstones, limestones, and many basalts would fall into this category. Preliminary telephone interviews with various geologists, petrologists, and experts in the area of rock weathering indicate that our proposed plan is both feasible and needed. With this encouragement, we are pursuing the matter further.

Preliminary contact on some potential resources for use in the physical models study (flumes at Colorado State University) have been developed.

Highlight Conclusions

The literature review of site characteristics, "Environmental Factors Affecting Long Term Stabilization of Radon Suppression Covers of Uranium Mill Tailings," was issued. Documents on rock durability and selection, and on channel flow and riprap design. "Long Term Survivability of Riprap for Armoring Uranium Mill Tailings and Cover: A Literature Review" and "Rock Riprap Design Methods and Their Applicability to Long-Term Protection of Uranium Mill Tailings Impoundments," are to be issued within a month. Experiment design for studies on rock selection procedures and physical models studies are underway.

Projected Work for Next Quarter

Subtask A, Baseline Data, will be completed by issuance of reports "Literature Review of Models for Estimating Soil Erosion and Deposition From Wind Stresses Acting on Uranium Mill Tailings," "Overland Erosion of Uranium Mill Tailings Impoundments: Physical Processes and Computational Models," and "Examples of Survival of Ancient Manmade Earthen Structures and Their Implications for Long-Term Stabilization of Uranium Mill Tailings." A task review and briefing is to be given to the sponsor at headquarters. Need, feasibility and study designs will be prepared and submitted for the physical models (flume), rock selection procedures, and soil deposition studies.

Reports Issued During the Quarter

"Environmental Factors Affecting Long-Term Stabilization of Radon Suppression Covers for Uranium Mill Tailings," L. W. Long and J. K. Young, NUREG/CR-2564. Pacific Northwest Laboratory, Richland, WA, March 1982.

TASK 2: INTERIM STABILIZATION OF TAILINGS

Task Project Manager: J. N. Hartley
Principal Investigators: M. R. Elmore, C. T. Li

FIN B-2370 (Task 2)

Objectives

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

Accomplishments During Report Period

We have acquired the use of a very suitable wind tunnel for use with the lab tests. Evaluation of the dust suppressants using the wind tunnel will comprise the majority of the lab tests. Conditions to simulate aging at an accelerated rate will be used in conjunction with the wind tunnel. A laboratory test plan outlining proposed activities and timetable has been prepared. Several more dust suppressant materials have been collected for the laboratory screening tests. Some additional suppliers have been located and are being contacted for samples.

Two mill sites were visited this quarter as potential hosts for the field tests. Both located in Utah, they were Atlas Corporation, Moab and Rio Algom Corporation, LaSal, Utah. Both mills are agreeable to the field test. More mills are to be visited in early April before a selection will be made. After review of equipment capable of applying dust suppressants to tailings at the field test, a system was purchased and has been received at BNW. It is a portable, gas-powered, airless sprayer--well suited to the requirements of the field test. A field test plan has been prepared and will be submitted to NRC for approval at the program review in April.

More material has been assembled for the state-of-the-art report and a draft version should be ready early next quarter.

A presentation was made in March to Bill Shaffer and Don Harmon, NRC personnel visiting at BNW. Objectives, milestones, progress, and future activities for this task were discussed.

Highlight Conclusions

Recent location of an available wind tunnel for the laboratory tests will greatly simplify testing and improve the reliability of test results. Screening tests should proceed rapidly with this major requirement being satisfied.

So far most mill operators appear willing to participate in the field test. This is very helpful in selecting the most promising site for the test. Other technical criteria can then be used to select it.

Projected Work for Next Quarter

The state-of-the-art report on tailings stabilization will be completed soon. Most of the laboratory screening tests will be completed, and selections made of the more promising materials to be tested in the field. The field test site will be chosen and preliminary site preparation, if needed, will be done. A weather station to monitor meteorological conditions at the test site will be selected and possibly installed at the site. A program review will be conducted at NRC headquarters in April.

Reports Issued During the Quarter

None.

Task 3. TAILINGS DEWATERING TECHNIQUES

Task Project Manager: R. W. Nelson

Principal Investigator: T. E. Gates

Support Investigators: P. G. Gutknecht, D. W. Mayer

FIN B-2370 (Task 3)

Study Objective

The objective of this study is to evaluate approaches to dewatering and drainage of tailings to reduce potential seepage into ground water and enable earlier and more satisfactory long-term placement of thick earthen covers over tailings burial pits. Special attention is being given to evaluating the linear consolidation analysis as a worst case estimation for evaluating settlement during tailings dewatering by under drains.

Accomplishments During Report Period

Subtask A. Evaluation of Linear Consolidation Theory Applicability

During the last quarter work on the evaluation of the applicability of linear consolidation theory, as applied to uranium mill tailings, continued by the consultants, Dunn and Anderson, commencing with a final summary report in mid-March.

Initially, consolidation of the tailings material results from two types of loading conditions:

- 1) weight of the tailings material
- 2) development of excess pore pressure from maintaining a drainage surface at the sides and bottom of the pit.

After deposition of the tailings has been completed and the tailings have become normally consolidated, additional consolidation will result from the placement of the overburden and cap material.

Deformation of the pile is of immediate concern for safe engineering management; just as significant, deformation strongly controls permeability variations within the tailings material. Hence, the distribution of fluid

velocities, which is critical for modeling chemical transport, is strongly influenced by the dynamics of the consolidation process.

Terzaghi's linear consolidation theory developed for the first time a procedure enabling engineers, based on mathematical rigors, to predict the time rate of settlement of structures constructed on fine grained soils. In addition, Terzaghi's theory isolated those variables which influenced and controlled the rate of settlement.

The basic assumed properties of his soil were:

- 1) isotropy of the material,
- 2) reversibility of stress-strain relations, under final equilibrium conditions,
- 3) linearity of stress-strain relations,
- 4) small strains,
- 5) the water contained in the pores is incompressible, and
- 6) the water flows through the porous skeleton according to Darcy's law.

One of the fundamental assumptions of the existing (Terzaghi's) theory of consolidation is that the capacity of the soil to transmit water during consolidation is a constant. This assumption that the coefficient of permeability is a constant is generally recognized as an over simplification of the physical process involved. The resulting errors arising from such assumptions will depend on the magnitude of the load increment and the resulting void ratio changes.

In considering a soil normally consolidating under its own weight, it will not have uniform soil properties throughout the depth. The variation of the consolidation parameters can be due to the following factors:

- 1) decreasing void ratio at greater depths due to higher consolidation pressures,
- 2) variation with depth of the soil itself due to nonhomogeneity, and
- 3) during sedimentation finer materials which settle first at greater depths.

Small strains occur in the "classical" method of analysis when the drainage distance, H , is assumed constant during consolidation. The resulting errors are negligible for problems of building settlement, where

only small strains can be tolerated, but for tailings which are highly compressible the error can be significant. Hence, to accurately model the consolidation process of tailings material the following needs to be considered:

- 1) large strain,
- 2) nonlinearity of stress-strain relations,
- 3) anisotropic soil, and
- 4) saturated and unsaturated flow.

Two additional parameters may need to be included; osmotic component, π , and plastic lag, η , however insufficient data exist at this time to quantify these terms and assess their importance on the consolidation process.

Interest in the analysis of deformation in partially saturated soils developed in the late 1950's with the work of Bishop (1959) and Aitchison and Bishop (1960). Using existing theory for saturated materials, these authors first attempted to extend the concept of effective stress to the zone of partial saturation. They proposed that effective stress, σ' , is equal to total stress, σ , less a weighted sum of U_w (pore water) and U_a (pore air). Thus,

$$\sigma' = \sigma - x U_w - (1 - x) U_a$$

with the weighting parameter x strongly dependent on saturation. The effective stress relation given above was later found inadequate to describe the volume change behavior of partially saturated soils.

Following an earlier suggestion by Bishop and Blight (1963) and the work of Matyas and Radhakrishna (1968), Fredlund and Morgenstern (1976) analyzed constitutive relations for unsaturated soils and on the bases of semi-empirical grounds showed that e (void ratio) and S_w (water saturation) had to be treated as functions of two independent stress state variables. Fredlund in 1979 suggested that the combination of $(\sigma - U_a)$ and $(U_a - U_w)$ as the most advantageous combination for describing consolidation of partially saturated soils.

Subtask B. Measurement of Tailings and Soil Consolidation Characteristics

No work was conducted under this task during the previous quarter.

Subtask C. Summary of Materials and Installation Procedures

The study of materials and installation procedures applicable to dewatering uranium mill tailings culminated in a report, "Review of Design Approaches Applicable to Dewatering Uranium Mill Tailings Disposal Pits." This report was issued as NUREG/CR-2608, PNL 4150, and a camera-ready copy sent to NRC in February.

This paper reviewed agricultural drainage theory as the most applicable for uranium mill tailings, although certain differences still exist in drainage objectives and the physical and chemical characteristics of the two drainage problems. These differences are summarized below, along with a recommendation for future research in each area.

- The differences between objectives of tailings dewatering and those of agricultural drainage must be considered. Agricultural drainage is employed to keep the water table at a depth favorable to crop growth. Tailings drainage is aimed at reducing the moisture content as much as possible to reduce potential seepage from the pit and enable the consolidation required for earlier cover placement to reduce radon gas emissions. Transient solutions should be applied to drainage of tailings pits with simulation of saturated and partially saturated flow to the drains.
- Consideration should be made for the alternating layers of slime and sand-type materials in the tailings that occur due to sluicing techniques. These layers will have differing permeabilities and saturation characteristics. Agricultural drainage theory employs assumptions that the media being drained is homogeneous.
- A prerequisite to the development of uranium mill tailings drainage design should be the careful consideration of potential construction problems. The construction of a dewatering system for a tailings pit is completely different than that of an agricultural drainage system. Agricultural drains and envelopes are placed in excavated trenches and covered with a few feet of soil. Tailings drains and sumps must be constructed before and during placement of tailings in the pit. Alternate drain layouts, such as multi-level systems, can be evaluated with modeling for benefits in drainage performance.

- The drains will logically exit to a single sump which will be pumped at defined intervals. Excess liquid required in tailings transport to the pit must be dealt with during filling and not allowed to fill the drain sump areas. Sedimentation or particles suspended in the moving liquid will settle in the sump and clog drain outlets to the sump, rendering the system inoperable. It is recommended that design, construction and operation procedures be developed to avoid this problem.
- Laboratory or field tests are recommended to establish the integrity of drainage system materials in a uranium mill tailings environment. Design of drains and envelopes should take into account the maximum allowable velocities through openings to prevent mineral deposition or other damage, the expected loadings and the chemical differences between agricultural and tailings drainage water.
- As tailings dewatering practice begins, it is recommended that systems in operation be evaluated for drainage performance with regard to drain discharge, material stability and construction methodology. Technology development can be aided by studying drainage design approaches and the resulting system performance.

Highlight Conclusions

Substantial progress has been made in assessing the applicability of the classical linear consolidation theory as applied to uranium mill tailings, in addition to defining the "state-of-the-art" for unsaturated soil consolidation.

Based on the drainage features unique to uranium mill tailings recommendations for future research are given. Transient solutions should be applied to describe liquid movement through saturated and partially saturated tailings. Modeling should be used to evaluate the benefits of drainage design approaches after careful consideration of potential construction problems.

Projected Work for Next Quarter

Work will be initiated on adapting the variably saturated flow code TRUST according to the methodology proposed by Fredlund and Morgenstern. The

task of modification will involve the replacement of the χ parameter subroutine, currently in the TRUST code, with a new subroutine in which the dependence of e and S_w on the two state variables are input either as a table or as a convenient polynomial function. The effect of layering and the changing soil parameters will also be considered.

If funding is approved and the capital items arrive during the next quarter, testing of the required input parameters for the TRUST code will be initiated.

Reports Issued During the Quarter

P. J. Gutnecht, T. E. Gates, "Review of Design Approaches Applicable to Dewatering Uranium Mill Tailings Disposal Pits." NUREG/CR-2608, PNL 4150. March, 1972.

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

Task Project Manager: G. W. Gee
Principal Investigators: D. R. Sherwood
R. J. Serne
Support Investigators: M. J. Mason
S. R. Peterson
M. E. Dodson
R. L. Erikson

FIN B-2370 (Task 4)

Study Objective

The objective of Task 4 is to assess the effectiveness, benefits and costs of neutralizing tailings to reduce the mobility of toxic and radioactive constituents under a full range of site and environmental conditions.

Accomplishments During Report Period

Treatment Methods Selection Review

Work continued on the literature review of treatment methods. Treatment techniques under investigation can be divided into three categories: neutralization treatments, encapsulation techniques, and specific ion removal techniques. These methods may be applicable to the problem of reducing contaminant mobility from uranium mill waste.

Neutralizing agents have been used in the treatment of acid mine drainage (AMD), which is similar in composition to sulfuric acid uranium mill tailings solution; thus, the technique and literature available on AMD should be quite valuable for U tailings studies. AMD comes primarily from the strip mining of coal and is formed by the bacterial oxidation of pyritic materials. Pyrite is an iron sulfide mineral (FeS_2) that oxidizes in the presence of air to form ferrous sulfate and sulfuric acid.

The acidic mine water formed during this oxidation process is capable of dissolving trace contaminants such as heavy metals common to pyritic formations.

Neutralization processes used for the treatment of AMD include:

Hydrated lime	(Ca(OH) ₂)
Limestone	(CaCO ₃)
Soda ash	(Na ₂ CO ₃)
Caustic Soda	(NaOH)
Combination Limestone-Lime	
Combination Alumina-Lime-Soda	

The cost of these processes are dependent on the composition of the acid mine water to be treated and can vary significantly. The greater the total acidity of the AMD the higher the cost of treatment as: High total acidity (low pH) increases the reagent usage required to achieve a neutral pH and also increases the amount of sludge (precipitant) formed during the neutralization process, which must then be emplaced in some type of disposal impoundment.

Contaminant encapsulation techniques identified to date may be divided into two types: asphalt products and cementing agents. Asphalt products and cementing agents probably are too costly to treat all of the uranium tailings generated in the milling process, but three scenarios have been identified in which these treatments could be effective.

- 1) At mill sites where tailings are being segregated into a slime fraction and a coarse fraction, encapsulation of the more hazardous slime material may be economical. This would only be the case if the less hazardous high volume sand fraction could be emplaced in mined-out ore pits or backfilled into abandoned mine shafts. The low cost of backfilling could off-set the high cost of encapsulation.
- 2) Encapsulation processes could also be used in the disposal of the neutralized sludge generated from a neutralization process.
- 3) Cementation may be economical for the treatment of alkaline tailings. Alkaline tailings that have a pH of 9 or higher would be compatible with most cementation processes which require a pH of 12 to set-up into a rigid structure. The amount of cementing agent required to achieve a pH value of 12 is much smaller than that required to raise the pH of acid tailings (pH approximately 2).

Specific ion removal techniques are those techniques that selectively remove an ion from solution. In the uranium mining industry specific ion removal has been used for reducing the soluble radium concentration in uranium mine water and tailings solution. Radium removal has also been performed in the treatment of some municipal water supplies. Radium removal is needed because of its very low discharge limit (30 pCi/l to unrestricted areas), and low maximum permissible concentration in drinking water (5 pCi/l). Three treatment methods have been utilized for the removal of soluble radium on a commercial scale. These are barium chloride treatment, cation exchange, and selective complexation. Study of the literature on these treatments is still underway.

Laboratory Analysis

Characterization of the solutions from Lucky Mc mill and the Exxon Highland Mill have been completed under the Leachate Assessment project. These solutions are being used to determine the effectiveness of neutralizing agents for the treatment of uranium mill tailings leachate. Total acidity analyses were done on the solutions to determine how tailings solution compares with acid mine drainage for acid content. Total acidity values for acid mine drainage are commonly in the range of 500 to 5000 milligrams per liter as calcium carbonate. The measured total acidity values for the Exxon Highlands tailings and the Lucky Mc tailings solutions were 4500 mg/l as CaCO_3 and 14,500 mg/l as CaCO_3 respectively. These total acidity values were used to estimate the quantity of neutralizing agent required to treat these solutions to a pH of seven in batch experiments. Batch experiments using the tailings solutions mentioned above and the neutralizing agents coal fly ash, calcium carbonate, hydrated lime, magnesia, soda ash, and caustic soda are nearly complete.

Highlight Conclusions

Literature collection and critical analysis for the May milestone document, "A Review of Tailings Treatment Techniques and Assessment of Their Applicability and Effectiveness for Typical U.S. Mill Sites," is nearly complete. Laboratory experiments on neutralization of acidic tailings solution continues.

Projected Work for Next Quarter

The completion of a topical report on treatment techniques applicable to the U.S. uranium industry is projected for next quarter. Results of the batch experiments and selection of the neutralizing agents to be used for additional testing will occur next quarter.

Reports Issued During the Quarter

None were issued.

TASK 5. DISPOSAL TECHNIQUES ALLOWING CONTACT WITH GROUND WATER

Task Project Manager - R. W. Nelson

FIN B-2370 (Task 5)

Study Objective

The principal objective of this study is to determine and evaluate those areas of potential environmental concern that may arise through disposal of buried tailings in contact with ground water. The study will include physical and chemical laboratory measurements, hydrologic flow evaluations of typical cases using laboratory measurements, and the associated leachate transport modeling of the typical cases to determine any long-term environmental effects in ground water. The typical cases will enable comparing seepage from above and below water-table pit disposal of tailings with pump-back drainage control.

Accomplishments During Report Period

Subtask A. Measurement of Tailings and Soil Characteristics
 M. E. Dodson and G. W. Gee

The principal work of this subtask is to determine, through laboratory measurements, the partially saturated water retention and permeability characteristics existing in soils and tailings at typical uranium mill sites.

Tailings and soils tested under this subtask are:

- Dawn Tailings
- Dawn Overburden
- Pathfinder (Lucky Mc) Tailings
- Pathfinder (Lucky Mc) Overburden
- Mancos Shale (Grand Junction, Colorado)

The following tests are completed for all five samples listed above:

- Particle density
- Particle size distribution
- Hydraulic conductivity at field density
- Air-dry water content
- Water retention characteristics at -300 cm, -1,000 cm, -3,000 cm and -5,000 cm

- pH of a saturated paste
- EC of saturated extract
- Percent organic matter
- Percent CaCO_3
- Cation exchange capacity
- Proctor testing (Mancos Shale and Dawn Overburden)

In addition, the water retention characteristics for Dawn and Pathfinder (Lucky Mc) tailings have been determined from -10 cm to -100 cm. Work on the determination of water retention characteristics from -10 cm to -100 cm for Dawn overburden, Pathfinder overburden, and Mancos Shale is nearing completion. An internal report will be prepared by May 1st summarizing the measured results.

Subtask B. Flow Analysis for Typical Tailings Pit Sections
P. R. Meyer

During the past quarter, the TRUST variably saturated flow code has been used to simulate three disposal pit scenarios. All cases were simulated as two-dimensional (vertical cross-section) systems. The results of these cases will help to analyze the flow system, determine contaminant transport from the pit, and determine the quality of ground water available to down-gradient water users.

The soil moisture properties (with the exception of the gravel) used in all three cases were previously determined from the Morton Ranch Uranium Mill site samples. The gravel data for use around the drains for all simulations was generated based on existing material property curves. With the completion of Subtask A, future cases will utilize soil properties from the additional mill sites.

The three simulations completed to date share a common geometry and construction design and represent a 600 foot trench or pit. The pit, originally dug in sandstone, is backfilled with 30 feet of overburden material. A three foot clay layer is installed over this, with a clay stub wall rising to 20 feet along the pit sidewall. Four gravel-enclosed drains spaced at 170 feet apart are assumed laid over the clay liner, and 85 feet of mine tailings are slurried into the pit. All simulations were initiated with tailings saturated to a height of 81 feet above the clay.

Conditions unique to each case are:

- Case 1 simulated a symmetrical half-pit problem situated above a horizontal regional water table which is ten feet below the clay liner.
- Case 2 considered the full pit, with the water table at the same level as in Case 1. The entire system, however, was tilted to represent flow associated with a regional ground-water gradient of 0.005.
- Case 3 differs from Case 2 only in that the regional water table was raised an additional 74 feet, so ground water flows into, and potentially through, the tailings pit.

The following results have been obtained from each of the three cases:

- Contour plots of moisture content and hydraulic potential within the modeled region at specified times.
- Time history of total water contained in the tailings material.
- Time histories of drainage rates and cumulative mass drainage from the pit.
- Time histories of outflow rates and cumulative mass outflow for the cases with regional ground-water movement.

Analysis of the results from Case 3, which use drains at a level below the regional water table, shows that water from the surrounding sandstone and lower overburden contributes heavily to the drainage fluid of the outer-most drains. Consequently, regional flow is affected, and a rather complicated flow system results during pumping from the drains. Although more water of better quality passes through the drains (approximately 2.5 times more enters the two drains near the cut off side wall liners), the results suggest that below water table tailings disposal may allow better overall control of the contaminated fluid than above water table disposal.

A good deal has been learned concerning the TRUST code itself. Insights into optimal grid design, time step control, and general simulation efficiency are being noted and will greatly aid future TRUST users.

Subtask C. Contaminant Transport Analysis of Typical Tailings
Burial Pits
A. E. Reisenauer. C. A. Newbill

During the quarter, analyses began on 1) the transport of leachates unaffected by pH, and 2) on the transport of acid leachates neutralized by the buffering capacity of the various soil materials used in the model cases.

The water flow results from the hydrologic analysis under Subtask B serve as input to these transport studies. For each typical case, Subtask B provides at each time step in the analysis a set of potentials for each model node and the flow between pairs of nodes. A set of velocity vectors are generated and then fluid pathlines are calculated.

In Case 1 (above the flat water table) insufficient leachate seeped through the clay to cause the water flow lines to reach the outflow boundary, when drains were adequately pumped.

In Case 2 (above the sloping water table) seepage reached the water table essentially only through the compacted overburden placed beneath the clay liner. Flow in the regional system then gradually migrated beneath the pit in the sandstone at a higher rate than the leachate in the overburden. Pathlines originating in the pit gradually moved upward toward the water table surface near the down gradient end of the pit and then migrated to the down gradient boundary of the flow system. Although water travel time from the pit to the boundary was only five years, the first leachate pathline to reach the same boundary took 71 years. Leachate from the far side of the pit took over 500 years to arrive at the down gradient boundary.

In examining the front of the acid leachate for the same pathlines, the amount of leachate that seeped out of the bottom only used up the approximate buffering capacity of the clay liner. Accordingly, essentially all of the contaminants that would be precipitated by acid neutralization will remain in the clay liner or very close to it in the buried overburden.

Case 3 indicated that all the leachate flowed into the drain or pumped sumps located near the stub sidewall liner. Essentially no leachate escaped to the water table. Upward seepage through the overburden and clay liner occurred at low rates due to the steep gradients resulting from pumpage of the drains above the clay side wall liner.

Subtask D. Ground Water Consequence Evaluation of
Buried Tailings Disposal
P. L. Oberlander

The consequence analysis for Case 1 was not required since the small leachate seepage which escaped the clay liner did not reach the outflow boundary in the level water table case. Also, seepage from the pit sides was essentially eliminated by the stub clay side wall liners and by pumping back from the drains in the tailings above the clay liner.

The Consequence Analysis for Cases 2 and 3 are currently underway but have not been completed.

Highlight Results and Conclusions

Results to date from comparing the below and above water table disposal have suggested two important ideas:

1. The control of leachate seepage for below water table disposal of tailings is both easier to accomplish and now appears to be more complete when using drains with seepage pump back.
2. The variety of seepage control strategies possible are much greater for below water table disposal. They are also much more involved to evaluate, due to the many alternate possibilities for leachate seepage control.

The first item is a natural consequence of greater possibilities for stronger and more complete control of below water table flow systems. Below the water table greater drawdowns are developed which give more definite control of the system energetics, hence better system control. However, above water table controls involve ever weakening gradients toward the under drains at the longer time associated with the final partially-saturated drainage from the tailings. Accordingly, the later above water table tailings seepage is less effective in removing leachate due to the weak energetics associated with the drains at longer drainage times.

Projected Work for Next Quarter

The measurements of materials and leachate properties for the other uranium mill sites in Subtask A will be completed early next quarter and will become available for use in setting up the subsequent cases.

Additional above and below water table cases will be set up and solved beginning with the flow systems under Task B. As each of the flow system results are obtained for the new case, Task C will be completed first to provide transport results and then Task D to provide the associated environmental consequences. With these results the relative benefits of above and below water table tailings disposal can be compared.

Reports Issued During the Quarter

None were issued.

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

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

FIN B-2370 (Task 6)

Objective

Assess measuring devices, techniques, and procedures for verifying acceptability of site and structure decontamination. Develop needed monitoring methods and equipment including testing and improvement for assessing release of radioactive and toxic materials from various facility components to allow establishing detailed guidance on monitoring requirements and strategies for evaluating compliance with 40 CFR 190.

The work embraced by this task is distributed among several subtasks. Each subtask toward which effort was applied during the quarter is identified and reported in the following pages.

Accomplishments During the Reporting Period

Subtask A. Field Comparison of Radon Progeny Measurement Techniques
(P. O. Jackson)

Objective

The objective of this subtask is to determine if there is a satisfactory, short-term measurement technique for the determination of chronic low-level exposures to radon progeny.

Accomplishments

During this quarter, it was necessary to continue to emphasize the clerical tasks of literature review, contacts with others doing research on the measurement of radon and radon progeny, and accumulation of information concerning available commercial instruments. Since capital equipment budget approvals were not received until the end of the quarter, it was not possible to procure

the instrumentation necessary for a laboratory evaluation or field evaluations of continuous radon and radon progeny equipment.

During the report period, technical liaison was established or expanded with research scientists at seven laboratories which are currently or have recently been involved with evaluation or use of equipment for continuous radon and radon progeny measurements at environmental exposure levels. We have requested an opportunity to participate in a round robin evaluation of grab sampling equipment for radon measurements organized by Dr. Isabell Fisenne of the Environmental Measurements Laboratory of the Department of Energy.

Because of the delay in procuring the capital equipment, it will not be possible to initiate laboratory or field evaluations for most short-term measurement techniques before mid-June 1982. Nevertheless, the experimental protocol for field measurements is being developed. One property owner in Edgemont, South Dakota has tentatively agreed to permit installation of continuous radon monitoring equipment in his home. Another owner of a vacant home located near the first has also tentatively agreed to permit installation of necessary equipment for program needs on a lease arrangement. These homes will be equipped to permit continuous radon and radon progeny measurements over a period of two to three months (and perhaps longer if necessary). The occupied house would also be equipped with continuous monitors of the condensation nuclei, relative humidity, temperature, and indoor air exhalation rate. The movement of air in the vicinity of the condensation nuclei will be detected using a sensitive, three-dimensional air motion detector. Outputs from these monitors will be recorded with a multichannel data logger to permit correlated studies. The vacant property will serve as a control, influenced primarily by external atmospheric conditions. A micrometeorology station will also be monitored with the data logger. This program is designed to determine which conditions and activities in the home most significantly affect radon and radon progeny concentrations for short and long-time intervals.

Projected Work for Next Quarter

Capital equipment items necessary for establishment of the radon/radon progeny experiments and other monitoring activities in the field program will be procured. A large volume, controlled atmosphere chamber, formerly used for

the exposure of animals to test atmospheres of ^{85}Kr , has become available to this program. This chamber will be equipped to permit the maintenance of a controlled atmosphere of radon and radon progeny as well as condensation nuclei and relative humidity and will facilitate the laboratory calibration and evaluation of equipment items which are too large for the small chambers previously constructed for the program.

Lease arrangements with property owners at Edgemont, South Dakota, will be finalized so that the field evaluation phase can begin as soon as the monitoring equipment is ready.

Reports Issued During the Quarter

None.

Subtask B. Yellow Cake Emission From Ventilation Exhausts

No activity to be reported for this subtask.

Subtask C. Critical Evaluation of Long-Term Radon Daughter Measurements and Needed Improvements (P. O. Jackson)

Objective

The objective of this subtask is to determine which, if any, of the available long-term radon and radon progeny measuring devices is the most accurate and cost effective.

Accomplishments

During the quarter, testing and evaluation of track etch detectors for long-term radon progeny measurements continued. Exposures were made in chambers at PNL, and at the PNL laboratory in Montgomery, Alabama. In addition, cross-calibration measurements were made in homes in Butte, Montana, and Edgemont, South Dakota.

Laboratory tests confirmed that a collimating filter cup type holder developed at PNL for Kodak Pathe' LR 115 film produced holes which were more easily distinguished under the microscope than films exposed without collimation. This was because collimation reduced the number of alpha particles which enter the film at shallow angles and which don't make clearly defined holes.

These devices were calibrated by using known radon exposures in homes in Butte, Montana. The calibrations there and at the EPA calibration facilities showed some variations in detector response that are still being investigated. However, when an average efficiency developed from the Butte, Montana exposures was applied to films from the Edgemont, South Dakota study, the resulting radon concentrations were more closely related to RPISU 100-hour radon progeny exposure measurements than were commercial track etch results for the sampling periods. The coefficient of variation of the ratio:

$$\text{pCi/l } ^{222}\text{Rn per Working Levels Exposure Rate}$$

decreased from 48% to 31% using the collimated cups. This study involved only 12 cups of each type in 12 homes in Edgemont, South Dakota, but it shows promise that existing methodology can be improved. Some of the variation in the ratio of radon to its daughters is caused by ventilation factors that differ from house to house. However, it appears that a portion may involve characteristics of the measurement system. When these devices are tested in the monitored homes described in Task A, it will be possible to determine the influence of environmental factors on the ratio, and the extent of its variation in those homes.

Projected Work for the Next Quarter

During next quarter, six RPISU units will be purchased for long-term evaluations. The interpretation of factors affecting track etch calibrations will continue. More laboratory test exposures will be made to determine the effect of ambient conditions on the detection systems. Correlative studies will be completed for long-term radon and radon progeny measurements made in Edgemont, South Dakota. Data from ongoing studies at other laboratories will be reviewed.

Reports Issued During the Quarter

None.

Subtask D. Critical Evaluation of Measurement Techniques for Uranium, ²²⁶Ra, ²²⁶Ra, ²³⁰Th, and ²¹⁰Pb in Effluents and Environmental Materials and Recommended Improvements (W. C. Weimer)

Objectives

The purpose of this subtask is to compare existing analytical procedures for quantifying U, ²²⁶Ra, ²³⁰Th, and ²¹⁰Pb in effluents and environmental materials for sensitivity. In addition, the individual sensitivities will be compared with regulatory standards; and where improvements are required, techniques will be extended or new techniques will be developed to produce methods which are sensitive enough to address the regulatory standards.

Accomplishments

The literature survey of required/proposed standards for environmental levels of the four radionuclides of interest was continued and was completed during this quarter. The literature survey of existing measurement methodologies was also completed during this quarter. The data from these two surveys were used to prepare a listing of those measurement techniques that have the feasibility of being used to quantify the four radionuclides at the levels of interest in environmental matrices. A brief interim report was prepared to summarize the findings of the two literature surveys.

Highlight Conclusions

Proposed limits for concentrations in soil appear to be at the lower limits of detection by currently available instrumental methods. Further work is needed to confirm this observation.

Projected Work for Next Quarter

The counting sensitivities and capabilities of all instrumental counting systems will be evaluated in the laboratory. The results of these studies will be used to assist in the determination of method cost effectiveness and in identifying areas where collection and analytical sensitivity improvements can be made.

Reports Issued During the Quarter

None.

Subtask E. Review Well-Logging Techniques (K. B. Olsen)

Objectives

This subtask's principal objectives are to evaluate and compare well-logging techniques to coring sample results, determine the effects of interferences (high natural uranium decay concentrations and variations of ^{40}K concentrations) on the well-logging instruments and recommend an acceptable procedure which can be applied to present and future remedial action programs.

Accomplishments

The literature review sub-subtask has been completed. In brief, the literature review determined that little, if any, work has been done using downhole detectors in remedial action programs. Considerable amounts of work have been done using the intrinsic germanium downhole detector for the location of uranium deposits in various areas of the United States and the Soviet Union.

A uranium tailings pile simulator has been constructed consisting of a 36-inch diameter cylinder layered with 18 inches of 0.78 pCi/g sand, 6 inches of 5.0 pCi/g tailings, and 18 inches of 0.78 pCi/g sand, respectively. This simulator has been used to determine if the detectors are capable of distinguishing between 5.0 pCi/g soil and 1.0 pCi/g background soils.

A request has been made to Silver King Mine of Edgemont, South Dakota, to case two holes through their tailings pile into the subsurface shale. An agreement has been reached where Silver King will supply Pacific Northwest Laboratories with core sample material from the cased holes. In addition, they will make available to us the trace metal analysis data from the core samples. Presently we are planning to make a field trip to Edgemont in the middle of April to do the profiling of the holes with both intrinsic germanium and NaI(Tl) detectors.

Highlight Conclusions

The literature search portion and the uranium tailings pile simulator tests using intrinsic germanium and NaI(Tl) detectors have been completed. Arrangements have been made with Silver King Mine to case two holes through

the tailings pipe and into the subsurface shale. A trip is anticipated in mid-April to profile these holes with both NaI(Tl) and intrinsic germanium detectors and compare these data with radioanalytical results from the core samples.

Projected Work for Next Quarter

To obtain data from the use of NaI(Tl) and intrinsic germanium detectors in the field by profiling two boreholes drilled through the tailings pile at Edgemont's Silver King Mine. These data will be intercompared from each of the detectors. An intercomparison of the well-logging results and the core sample results will be examined. In addition, field comparisons of the efficiency, reliability, practicality, cost-effectiveness of each of the methods will be evaluated. The above data will also be the initiation of sub-subtask 2 (Determination of Expected Field Interferences from High Natural Uranium Decay Concentrations and the Variability of the ^{40}K Concentrations in the Test Soil).

Reports Issued During the Quarter

None.

Subtask F. Evaluate and Recommend Sampling Strategies for Temporal and Spatial Evaluation of Contamination (V. W. Thomas)

Objectives

The objectives of this research is the development of guidelines which will allow the detection and evaluation of radioactive contamination spread from uranium tailings piles which may extend up to several kilometers. Following these guidelines will give a reasonable certainty that spread of contamination will be detected by defining the number, distribution, and timing of measurements required to achieve this degree of certainty while minimizing the magnitude of the effort required.

Accomplishments During the Report Period

This subtask was initiated during the quarter. Literature information relevant to measurements of contamination spread from tailings piles was and is being collected for review. Among others, these include radiological surveys of inactive uranium mill sites, most of which are included in the UMTRA

Program. These surveys were conducted by the Environmental Protection Agency in about 1975; again by Ford, Bacon, Davis, Utah in about 1977; by Oak Ridge National Laboratory in about 1980; and engineering assessments were done by Ford, Bacon, Davis, Utah in 1980. Radiological surveys have been performed at formerly utilized MED/AEC sites in connection with FUSRAP. We are also evaluating the experience and data generated during the Edgemont, South Dakota, Remedial Action Program (FIN B-2217). We are also coordinating our efforts with those being extended in the Environmental Clean-up Standards for Low-Level Radioactive Waste Materials Project (FIN B-2216). Evaluation of other related programs for inclusion in the data base or statistical techniques include the following studies: An Environmental Study of Active and Inactive Uranium Mines and Their Effluents, Parts 1 and 2, J. G. Droppo, et al., PNL-3069, Survey of Radionuclide Distributions Resulting from Churchrock, New Mexico Uranium Mill Tailings Pond Dam Failure, W. C. Weimer, et al., NUREG/-2449, PNL-4122; A Field and Modeling Study of Windblown Particles from a Uranium Mill Tailings Pile, L. C. Schwendiman, et al., NUREG/CR-1507, PNL-3345; Monitoring for Compliance with Decommissioning Termination Survey Criteria, C. F. Holloway, NUREG/CR-2082, ORNL/HASRD-95; and Technology and Costs of Termination Surveys Associated with Decommissioning of Nuclear Facilities, J. P. Witherspoon, Oak Ridge National Laboratory, NUREG/CR-2241, 1981.

The assistance of a PNL statistician has been enlisted for the required statistical analyses (R. R. Kinnison). He has commenced evaluation of data from the above reports for inclusion in the statistical data base. Telephone or personal conversations have been made with other researchers familiar with radiological survey procedures.

Projected Work

Collection, evaluation, and comparison of previous survey data for inclusion in the statistical data base will continue.

Reports Issued During the Month

None.

Subtask G. Sampling Techniques for Detecting Contamination of Surface and Ground Waters by Tailing Leachate (W. C. Weimer)

Objectives

The objectives of this subtask are the evaluation of existing standard techniques for measurement of levels of contaminants in natural waters for application at uranium recovery sites. Known dissolved contaminants that are potential hazards to water quality which will be considered in this study include the radioactive uranium daughter products (in particular thorium and radium) and stable elements Se, As, Sb, Zn and Cu.

Accomplishments During the Report Period

This task was initiated during this quarter. As part of this initiation, the assistance of a PNL statistician (R. R. Kinnison) has been enlisted. His function will be to evaluate the statistical aspects of previous ground-water survey programs which have been performed around sites where hazardous materials have been dumped or buried. From the evaluation of these previous programs, he will develop a sampling strategy and protocol for both surface waters and ground waters near uranium recovery sites.

A literature survey has been started to gather data regarding the sampling plans that have been used to monitor surface water and ground water contamination at various sites. Thus far, it appears that relatively little information is available from these past studies. We have also completed the review of the analytical method capabilities for measuring dissolved/particulate water contaminants.

Highlight Conclusions

The statistical aspects of both ground water sampling and surface water sampling will be a major consideration to be addressed by this subtask. These may far outweigh any analytical deficiencies that we uncover.

Projected Work for Next Quarter

Both the literature review of analytical methodologies and the literature review of previous ground water sampling plans will be completed. A laboratory determination of analytical interferences in the measurement methodologies

will also be completed. A statistical evaluation of the sampling plans will be started.

Reports Issued During the Quarter

None.

TASK 7. TECHNICAL PROGRAM MANAGEMENT

L. C. Schwendiman

FIN B-2370 (Task 7)

Objective

The purpose of this task is to provide the program management function of all the uranium recovery waste management studies contracted to PNL from the Waste Management Branch, NRC-RES. Principal functions are to coordinate the efforts of scientists and engineers assigned to tasks, to prepare and issue quarterly technical and administrative reports, to 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 Reporting Period

Close liaison was maintained and numerous contacts made with NRC project managers and task managers and their principal investigators. Some of the particular accomplishments were the following.

- Acquisition of project and task development plans for the Uranium Recovery studies at PNL. Although primarily an internal project management tool, these plans will be made available to NRC project managers.
- Revision of the 189 for FY-1982 for FIN B-2370, FIN B-2292, and B-2267, reflecting changes required by the work authorization letters. These revised 189's will be transmitted to NRC in April. Progress was made toward writing the FY-1983 proposals.
- Monthly administrative reports were submitted to NRC for January, February, and March.
- A trip was completed which included the following contacts:
 - February 5 EG and G, Idaho Falls, Dennis Goldman et al.
 - February 8 Rogers and Associates Engineering Corporation, Salt Lake City; Vern Rogers and K. K. Nielson
 - February 10 Sandia Laboratories, Albuquerque; Harold Rarrick , Paul O'Brien

February 10 UMRAP Office, DOE, Albuquerque; M. L. Matthews
February 10 Noel Savignac, Albuquerque, NM
February 11 New Mexico Environmental Improvement Division, Al Topp, et al.
February 12 Los Alamos National Laboratory, Los Alamos, NM; Dave Dreesen,
et al.

The purpose of these visits was to determine the status of related work at these installations and to inform those visited of the scope and objectives of PNL studies. A secondary objective was to consider capabilities of some individuals for peer reviews of PNL research.

- A possible conflict of interest issue was settled in a conference call with NRC-NMSS and RES principals and staff of PNL on March 19. A memo was distributed confirming the conclusions reached.
- A program review was planned and conducted for Don Harmon (RES) and Bill Shaffer (NMSS) on March 2 and 3 at Richland, Washington. A program review to be held April 20 at the Willste Building, Silver Spring, MD, was planned. Long-term stabilization with riprap and interim stabilization research will be reviewed.
- Capital equipment requests and priorities were submitted for the PNL tasks.
- Near-final drafts of several PNL reports were read and comments furnished the authors.
- The first quarterly progress report for FY-1982 was assembled and issued in January 1982.

Highlight Accomplishments

Administrative reports were submitted in timely fashion. Revisions of three 189's for FY-1982 were readied for sending to NRC-RES. The information exchange trip was successful. A possible conflict of interest issue was resolved.

Projected Work for Next Quarter

- New 189's for FY-1983-1984 will be prepared and submitted.
- A program review of Tasks 1 and 2 of FIN B-2370 will be held in Silver Spring on April 20.
- A program review of Tasks 3, 4 and 5, FIN B-2370 and FIN B-2292 will be held in Silver Spring the last part of May.
- Project and Task Development Plans will be completed and/or updated to reflect changes in scope and direction resulting from feedback from program reviews and task progress.
- Several documents scheduled to be completed will be reviewed prior to submission.
- External peer reviewers will be subcontracted for reviewing segments of the uranium recovery research underway.

Reports Issued During the Quarter

L. C. Schwendiman, et al., "Uranium Recovery Research Sponsored by the Nuclear Regulatory Commission at Pacific Northwest Laboratory." Quarterly Progress Report. PNL-4105-1, January 1982.

ASSESSMENT OF LEACHATE MOVEMENT FROM URANIUM MILL TAILINGS

Task Project Manager: G. W. Gee

Principal Investigators: R. J. Serne
R. W. Nelson
S. R. Peterson

FIN B-2292

Study Objectives

The principal objectives of this program are to develop experimental and computer modeling tools to assess the long-term environmental impact of leachate movement from uranium mill tailings.

Accomplishments During Report Period

Project Management (01)

During this quarter, a draft of the 189 for FY83 was made. The 189 for FY82 has been accepted by NRC after the slight updating that was done this quarter. A Project Planning Document was also written to give a detailed breakdown of work activities and to set dates for the completion of various work assignments.

A presentation on the current status and future plans regarding Leachate Assessment was made to Don Harmon and Bill Shaffer on March 2 and 3. No particular issue was raised in the discussion except the need for a clearer statement of the relationship between this work and B-2370, Tasks 3, 4, and 5.

Laboratory Analysis and Geochemical Modeling (02)

Characterization of the additional tailings solutions collected (hereafter referred to as "new") was completed (Table 3) and compared with tailings solutions previously sampled (hereafter designated "old"). All analyzed constituents except for iron in the "old" and "new" Highland Mill (H.M.) tailings solutions were approximately equivalent in concentration. Total Fe

concentrations in the "old" H.M. tailings solutions were about 2200 ppm while those in the "new" solutions are on the order of 950 ppm. The difference in total iron content can be explained, in large part, by changes in mill processing procedures and by a change in the sampling location.

Depending on the amount of uranium in the ore, varying quantities of Fe are used in the extraction process. The "old" samples were collected at the spot where the pipe leading from the mill to the tailings pond discharges into the pond. The "new" tailings solution was taken near the edge on the north side of the present tailings pond, and had a higher pH and Eh than the "old" tailings solution. The "old" tailings solution had an Eh and pH of 625 millivolts and 1.8 respectively, while the comparable values for the "new" solution were 750 and 2.2 respectively. Assuming an amorphous ferric hydroxide is controlling the Fe^{3+} activity in solution, the Fe activity would decrease by over an order of magnitude in going from pH 1.8 to a pH of 2.2. The higher Eh would ensure that more of the iron existed in solution as Fe^{3+} , enabling it to precipitate with the relatively insoluble Fe(III) oxy-hydroxides (such as amorphous ferric hydroxide).

The new Lucky Mc tailings solution has a higher total Fe content and significantly lower concentrations of Na and Mg than the "old" Lucky Mc tailings solution. These differences are, again, probably due to changes in the amounts of chemicals used in the processing of the ore and the lower pH and Eh values encountered in the "new" tailings solution. See Table 4 for the chemical analysis of the "new" Lucky Mc tailings solution.

Eh, pH, and selected radionuclide determinations were finished for all samples taken from the Lucky Mc evaporation pond #4 (Table 5). Seven of these samples have been selected for additional characterization (Table 5). This characterization will include amorphous stripping and chemical analysis of the stripped assemblage to quantify which contaminants are associated with particular solid phases. The samples chosen for additional characterization span the range of conditions that exist beneath the evaporation pond. This pond was drained in the summer of 1980, covered about 90

acres, and had been in operation for 4 years at the time of drainage. The average fluid level maintained during the active disposal period was 12 feet. All samples taken below a depth of approximately 12 inches were of near neutral pH, which indicates the capacity of the sediment to buffer the low pH tailings solution. The concentrations of the measured radionuclides decreased with depth illustrative of the higher pH with increasing depth and the adsorptive capacity of the soils.

During this quarter, initial work was also completed on the selection of analytical methods for the determination of uranium mill tailings chemistry. Table 6 lists elements important in the chemistry of uranium mill tailings along with the proposed analytical technique for each element. In general, inductively coupled plasma (ICP) is suggested for macro-cation analysis while graphite furnace atomic absorption (AA) is recommended for trace metals. We have demonstrated with the AA that we can meet MPC or lower concentrations for As, Cd, Cr, Cu, Fe, Mo, Pb, Mn and V. Se may require hydride generation to get below 0.05 ppm (0.01 ppm is the MPC) but continued efforts may lower Se detection limits. Efforts in assessing the comparative analytical capabilities of AA and ICP for various elements are continuing. Important anions will in general be determined by ion chromatography (IC) and radionuclides by the counting techniques identified in Table 6.

A geochemical model was selected for doing the ion-speciation-solubility and mass transfer modeling projected for the second half of FY82 and during FY83. Documentation for this model, which has been developed at Battelle, Pacific Northwest Laboratories, will be published in July of 1982. Verification work was begun this quarter in comparing this resident PNL model to existing computer codes such as PHREEQE (Parkhurst et al., 1980), WATEQ3 (Ball et al., 1981) and EQUILIB (Morrey, 1977). Although the PNL code gives the same answers as these existing codes for the ion-speciation-solubility and mass transfer capabilities that have been tested, the PNL code has four advantages. These include: 1) a larger, more thoroughly documented data base, 2) greater ease in using the mass transfer capabilities (more user oriented), 3) better convergence capabilities on a wide range of input data and 4) faster convergence in the cases tested so far.

The acid attack permeability cells leached with uranium mill tailings for about 3 years were dissected. The hydraulic conductivities of these cells over time are now being determined. The permeability cells leached with synthetic tailings solution are still on-going and will be monitored for changes in the permeability.

Hydrologic and Mass Transport Modeling (03)

Several modifications were made to the original grid generation program because it outgrew the size capacity of the PDP 11/55. Among these were:

- 1) An overlay version of the program called DIGRD that overlays 10 sub-routines and provides room for additional subroutines.
- 2) A subroutine that generates a 1/2" by 1/2" grid on the digitizing table. A matrix relates grid elements to table grid, thus facilitating the recall of a particular element and elemental information.
- 3) Several grid editing options including,
 - node positioning within an element
 - node and element connectivity deletion
 - File-Q compression
 - element connectivity creation
 - interface re-positioning
- 4) A separate program which reads the TRUST input files and formats a save file to be read by DIGRD.

A user's manual for the DIGRD program has been prepared in draft form and is under review. The final code for the "interface re-positioning" edit node is to be developed in the next quarter.

A request was made to Harwell for information on the MA codes (MA 28 and 32), which are efficient matrix solvers. These solvers are currently employed by Lawrence Berkeley Laboratory in a matrix version of the TRUST code.

Highlight Conclusions

Characterization on the additional tailings solutions was completed, and state-of-the-art methods developed for the chemical analysis of uranium mill tailings solutions. A presentation on leachate assessment was made to the NRC. A geochemical model for use on the chemical modeling portion of this study has been selected and verification of the model has begun.

Projected Work for Next Quarter

Detailed procedures will be written regarding analytical techniques for the determination of the constituents of high ionic strength tailings leachates. Thermodynamic data on trace elements important to uranium mill tailings but not found in current data bases will be added to the resident PNL computer model. All needed characterization work on tailings, tailings solutions, plant process water, groundwater and sediments will be completed. Chemical modeling of tailings solutions and the sediment amorphous stripping techniques will be initiated. Column leaching experiments performed with spikes of radioactive trace metals will also begin next quarter.

Reports Issued During the Quarter

None

References

- Parkhurst, D. L., D. C. Thorstenson and L. N. Plummer. 1980. "PHREEQE - A Computer Program for Geochemical Calculations." USGS/WRI-80-96, 216 pp.
- Ball, J. W., E. A. Jenne, and M. W. Cantrell. 1981. "WATEQ3 - A Geochemical Model with Uranium Added." USGS Open File Report, 81-1183, 81 pp.
- Morrey, J. R. 1977. "Equilib - A Computer Program to Predict Complex Aqueous Equilibria from 0 to 300 C." BN-SA-1297, Battelle Pacific Northwest Laboratories, Richland, Washington, 24 pp.

TABLE 3. "NEW" EXXON HIGHLAND MILL TAILINGS SOLUTION,
Average of 3 Analysis(a)

Element	ICP, mg/l	AA, µg/l	IC #1, mg/l(b)	IC #2, mg/l(b)
Ag		50		
Al	405.5 ± 2.12			
As		425.2 ± 40		
B	<1			
Ba		<50		
Ca	548 ± 16.97			
Cd		38.2 ± 2.62		
Cr		1412 ± 18.6		
Cu		1.10 ± 0.02		
Fe	947.5 ± 2.12			
K	39.5 ± 0.0			
Mg	495.5 ± 0.71			
Mn	44.0 ± 0.0			
Mo		<50		
Na	332 ± 2.83			
Pb		<50		
Se		1.18 ± 0.03		
Si	208.5 ± 0.71			
Sr	8.0 ± 0.0			
V		10.58 ± 0.63		
F			4.05 ± 0.07	2.50 ± 0.0
Cl			291 ± 24.04	
NO ₂				
NO ₃			7.0 ± 0.0	
SO ₄			7920 ± 70.71	8830 ± 0.0

(a) pH = 2.24; Eh = 750 (mv); Error band is one standard deviation to either side of mean.

(b) Two independent laboratories ran the anions by ion chromatography.

TABLE 4. "NEW" LUCKY MC TAILINGS
SOLUTION Averaged^(a)

	<u>ICP, mg/l</u>	<u>AA, µg/l</u>	<u>IC #1, mg/l^(b)</u>	<u>IC #2, mg/l^(b)</u>
Ag		< 50		
Al	1029.5 ± 6.36			
As		19.33 ± 0.63		
B	1.2 ± 0.28			
Ba		92.0 ± 4.2		
Ca	604.5 ± 26.2			
Cd		281.5 ± 16.3		
Cr		2357.5 ± 38.3		
Cu		1.51 ± 0.09		
Fe	2778.5 ± 10.61			
K	156 ± 5.7			
Mg	1222 ± 58.0			
Mn	162.5 ± 3.54			
Mo		8412.5 ± 203		
Na	1632.5 ± 10.61			
Pb		874.5 ± 22		
Se		1.61 ± 0.05		
Si	283 ± 11.31			
Sr	14.0 ± 0.0			
V		14.45 ± 0.47		
F			28 ± 0.0	30 ± 0.0
Cl			1090 ± 0.0	1300 ± 0.0
NO ₂				
NO ₃			302 ± 7.07	313 ± 0.0
SO ₄			26395 ± 445	27100 ± 283

(a) Error band is one standard deviation to either side of mean.
pH = 1.22; Eh = 698.

(b) Two independent laboratories ran the anions by ion chromatography.

TABLE 5. PROFILES OF LUCKY MC EVAP. POND #4 (pCi/g)

Location	Depth	pH	^{210}Pb (pCi/g)	^{238}U	^{230}Th	^{226}Ra	Eh
7400	0-2"	3.31	1747	6.74	15,363	368	590
	3-6"	3.43	1240	10.75	11,507	382	593
	*8-10"	4.92	25.4	218	4,320	108.	560
	12-15"	7.80	17.35	9.44	106.5	10.2	432
	16-20"	7.78	3.49	6.39	20	7.19	441
9+00	0-2"	3.46	1715	37.56	9743	518.7	545
	3-6"	3.38	13.29	37.1	2406	54.3	591
	10-12"	7.43	8.83	36.7	88.8	9.43	468
	15"	7.48	2.78	18.1	33.60	10.64	460
Pipeline	0-2"	3.84	1380	32.2	7303	372	545
	3-6"	4.46	688	53.2	8517	154.3	520
	*10-12"	6.25	95.5	470.	3174	144.7	405
	15-17"	7.60	2.35	51.94	< 20	16.31	344
	*24"	7.80	1.60	7.86	< 20	7.12	345
NE Corner	0-2"	3.65	1285	32.8	123,331	408	478
	3-6"	3.70	980.6	12.13	12,579	213.7	485
	6-10"	4.27	17.46	66.49	3,285	51.1	626
	*12-15"	6.78	4.43	163	266.2	48.	433
	24"	7.70	30.3	18.2	294.6	15.	405
NE Side	1-3"	4.02	477	28.9	8703	125.6	621
	12"	8.00	1.66	12.6	20	6.14	442
	18"	7.15	2.17	5.71	20	3.78	427
	24"	7.15	3.46	6.96	21.5	5.15	423
Bulk	*White Salt	3.62	474	17.4	4,419	140	568
	*Layered						
	Surface #1	2.95	1026	11.87	15,235	279	610
	*Layered						
	Surface #2	2.97	1170	24.24	15,985	296	610
	*Red Sand #1	3.91	53.3	54	4,030	45.5	684
	*Red Sand #2	3.83	57.4	176	4,111	41.2	697

* = Samples selected for additional characterization.

TABLE 6. ELEMENTS/ANIONS CONSIDERED AND PROPOSED ANALYTICAL METHODS

Elements/Anions	ICP	Ness	IC	Titrate	AA	α -Spec	Intrinsic
<u>Macro Cations</u>							
Al	X						
B	X						
Ca	X						
Fe	X						
Mg	X						
Mn	X						
Si	X						
Na	X						
Sr	X						
NH ₄		X					
<u>Trace Metals</u>							
As					X		
Ba					X		
Cd					X		
Cr					X		
Cu					X		
Pb					X		
Mo					X		
K					X (a)		
Se					X (b)		
Ag					X		
V					X		
Zn					X		
<u>Radionuclides</u>							
²¹⁰ Po						X	
²¹⁰ Pb							X
²³⁰ Th							X
²²⁶ Ra							X (c)
U							X
<u>Anions</u>							
NO ₃			X				
PO ₄			X				
SO ₄			X				
Cl			X				
F			X				
HCO ₃ -CO ₃				X			
(a) AA flame							
(b) AA hydride							
(c) intrinsic/laser fluor							

RADON AND AEROSOL RELEASE FROM
OPEN PIT MINES

Project Manager:	R. W. Perkins
Principal Investigator:	V. W. Thomas
Support Investigators:	N. A. Wogman, M. L. Mauch*
	K. K. Nielsen*

FIN No. B-2279

Study Objectives

This investigation has the principal objective of determining the quantity of radon released per unit of uranium produced from open pit mining. A secondary objective is to determine the nature and quantity of airborne particles during mine operations. Models for estimating radon release are to be developed and tested.

Accomplishments During this Report Period

The final report for this project "Radon and Aerosol Release From Open Pit Uranium Mining" NUREG/CR-2407, PNL-4071 has completed the second phase of internal review. The camera ready copy has been prepared and, as requested by NRC through L. C. Schwendiman, a xerox copy of the camera ready copy was given to Don Harmon, March 3 to be delivered to George Birchard in Washington, D. C.

Highlight Conclusions

A copy of the final report was delivered to George Birchard in March 1982

Projected Work for Next Quarter

Appropriate comments from George Birchard will be incorporated into the final report, the camera ready copy will be forwarded to NRC and copies of the final report will be sent to the participating mines.

*Presently at Rogers and Associates Engineers, 515 E 4500 So., Suite G100, Salt Lake City, Utah 84107.

Reports Issued During the Quarter

Final report "Radon and Aerosol Release From Open Pit Uranium Mining",
NUREG/CR-2407, PNL-4071, V. W. Thomas, K. K. Nielson, and M. L. Mauch,
March 1982.

ATTENUATION OF RADON EMISSION
FROM URANIUM MILL TAILINGS

Project Manager: R. W. Perkins
Principal Investigator: W. B. Silker
Support Investigators: V. C. Rogers,* K. K. Nielsen*
Rogers and Associates Engineering Corporation (RAE)
E. A. Lepel and D. R. Kalkwarf, PNL

FIN B-2267

Study Objectives

The objective of this research is to define straightforward and 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 for measuring their effectiveness after emplacement so that a regulatory guide can be developed and NRC's responsibilities to ensure safe tailings can be met.

Accomplishments During the Report Period

The principal investigator, W. B. Silker, is retiring from PNL on 30 April 1982, and direction of this program is being assumed by Dr. D. R. Kalkwarf. Sufficient time has been allocated to assure an orderly transition of programmatic responsibilities so that work will continue according to the planned schedule. W. B. Silker has agreed to remain available in a consultant capacity to provide assistance as needed to ensure timely completion of this research effort.

Task 3. Select and Test Candidate Cover Materials.

A report entitled "Radon Attenuation Properties of Candidate Cover Materials," is in final draft and is receiving internal review. After this review, and PNL clearance, a camera ready copy will be forwarded to NRC for printing and distribution.

Task 4. Field Confirmatory Studies

A report entitled "Comparison of Radon Diffusion Coefficients Measured

in the Field to Laboratory Measured Values" is also in final draft and will be subjected to internal review. Its publication as a NUREG document is also imminent.

Task 6. Alternative Measurement of Diffusion Coefficient (RAE)

The remaining transient radon diffusion coefficient measurements were completed during this quarter on the comparison soils furnished by PNL. Additional measurements will be conducted by PNL to more closely approximate the conditions of moisture and compaction tested by RAE, however, reasonable agreement in determined values for the diffusion coefficients by the two methods were obtained.

In addition, the radon diffusion coefficient was measured for a column of uniform glass balls (4 mm diameter), and for a column filled with vertical cylinders. The cylinders were to simulate large vertical diffusion channels without the complicating influence of tortuosity. The glass balls also had relatively large diffusion channels, but also had a well-defined tortuosity.

Table I illustrates the measured radon diffusion coefficients for the reference materials. As indicated, these diffusion coefficients should be corrected for atmospheric pressure before comparing with other data. The diffusion coefficient for glass balls equals those for air when the tortuosity (0.68) from Equation 2 is considered. As indicated, a relative standard deviation of 13% was observed for the dry Dunite measurements. This is considered to be a typical precision for the determination of a diffusion coefficient except in cases of nearly-saturated soils, where sample preparation techniques dominate the experimental uncertainty.

The determined values of $0.061 \text{ cm}^2/\text{s}$ for Dunite sand should be compared with a value of $0.063 \text{ cm}^2/\text{s}$ reported by PNL, and the values for air are in agreement with those reported in the literature.

The draft final report on the radon diffusion measurement is in final preparation and will be sent to PNL in early April.

Task 7. Modeling (RAE)

The draft report on the radon diffusion model previously submitted to PNL was revised for final distribution. The revisions included responses to comments by PNL reviewers as well as a more detailed description of

model parameters, particularly tortuosity.

The notation used for radon flux was altered for consistency with previous NRC reports. In order to provide a more clear understanding of

TABLE I

RADON DIFFUSION COEFFICIENTS* MEASURED FOR STANDARD DRY MATERIALS

<u>Sample</u>	<u>Porosity</u>	<u>D(cm²/s)</u>	<u>Sample</u>	<u>Porosity</u>	<u>D(cm²/s)</u>
Air	1	0.110	Dry Dunite	0.46	0.050
Air	1	0.120	Dry Dunite	0.45	0.056
			Dry Dunite	0.45	0.066
Glass Balls	0.33	0.078	Dry Dunite	0.45	0.068
Cylinders	0.25	0.124	Dry Dunite	0.45	0.064
					0.061 $\pm$ 0.008

* Measured at 0.9 atm. pressure. Multiply by 0.9 to correct to standard conditions.

the various radon diffusion coefficients used in the report, additional illustrations were also included to provide conceptual definitions. Other equations were revised to utilize standard notations for the fundamental constants used by the model, such as the diffusion coefficients in pure air and pure water.

In order to more explicitly define the effect of tortuosity in radon diffusion, this parameter was also defined in the revised report, and was included as an explicit parameter in the radon diffusion model. It was recognized that tortuosities are not generally measurable, and that their values must be estimated. Although natural soils are often well represented by the constant value = 0.66, a more detailed calculation was used in the present model to accommodate the wide range of compactions and grain sizes being considered. Tortuosity is primarily a function of the soil porosity and of the width of its pore size distribution. The determination of was partially based on considerations of uniform packed spheres, from which the following relationship with porosity was derived:

$$\tau = \left\{ 1 + \left[\frac{3(1-P)}{2\pi} \right]^{2/3} \right\}^{-1} \quad (1)$$

This equation accounts for the less-tortuous diffusion pathway which occurs when uniform spheres are less-densely packed, and reaches the most tortuous value of 0.66 for the close-packed geometry when $P = 0.26$.

When spheres of different sizes are mixed, small spheres may fill voids between larger spheres. In close-packed geometry, a binary mixture of very large and very small spheres would have the tortuosity of each size superimposed, resulting in a combined tortuosity of $= (0.66)^2$. If three very different sizes were mixed, the exponent could become 3, or in general, equal to the width of the grain size distribution normalized to the ratio of inscribed sphere radii. This ratio is 11.3 for close-packed spheres, and using the grain-size to pore-size model of Arya and Paris,⁽²⁾ becomes 44 if the width of the pore-size distribution is used instead of that for the grain size distribution. The resulting tortuosity expression used in the diffusion model calculations was

$$\tau = \left\{ 1 + \left[\frac{3(1-P)}{2\pi} \right]^{2/3} \right\}^{-(1+2GSD/44)} \quad (2)$$

where

GSD = geometric standard deviation of the pore-size distribution.

The illustrations of calculated diffusion coefficients were revised to utilize the above expression, and the diffusion model report was returned to PNL for final clearance.

Task 8. Handbook (RAE)

The handbook entitled, "A Handbook for the Determination of Radon Attenuation Through Cover Materials," NUREG/CR-2340, was distributed early in this quarter. After further discussions with PNL and NRC, it was decided that revisions to this handbook at the present time would be premature. Other parameters, such as advective transport of radon, should potentially be included in such revisions, but have not yet been characterized because this research is just now beginning. Revisions will be considered in late 1982.

Task 10. Validation Studies with Tailings Stabilization

Samples of three soils, a Mancos shale, a Bentonite clay and a local adobe clay have been obtained from Grand Junction, Colorado. These materials were used, singly and in combination as tailings covers at the DOE-UMTRAP test site. These soils are currently being evaluated with the RATF to determine the response (of the rate of radon diffusion) to variations in soil moisture and compaction. These data will be used to predict radon flux attenuation under conditions of the applied cover which will then be compared with flux reductions determined by field measurements.

Highlight Conclusions

Continued effort to model radon attenuation from basic physical concepts including a factor for tortuosity and laboratory experiments gave confidence that the physical processes of radon diffusion are well understood. A handbook for determining radon attenuation was issued.

Projected Work for Next Quarter

Reports of subtasks on radon attenuation in candidate materials, comparison of radon diffusion coefficients in laboratory and field measurements, and radon diffusion models will be completed and issued. Studies on advection as a factor in radon attenuation will be initiated. Confirmatory larger scale field studies will be planned.

Reports Issued this Quarter

"A Handbook for the Determination of Radon Attenuation in Cover Materials," NUREG/CR 2340, RAE-18-1, PNL-4084 was issued and distributed.

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

Project Manager: R.J. Serne

Principal Investigators: W.J. Deutsch and N.E. Bell

Support Investigators: B.W. Mercer, J.F. Shade and S.E. Petty

FIN B-2379

Study Objectives

The purpose of this study is to evaluate in-situ uranium mining operations and restoration procedures in order to provide information to NRC for the purpose of establishing regulatory guidelines for the environmentally-sound operation and abandonment of these mines. In addition to an evaluation of existing production/restoration procedures, assessments will be made of the monitoring systems in use and of the potential for deleterious hydro-geochemical alterations resulting from lixiviant excursions and ore-zone mining.

Accomplishments During Reporting Period

Task 1: Project Management

At the request of NRC, this project was reviewed at NRC headquarters in Silver Springs on February 3rd. Project personnel discussed the objectives, scope, accomplishments and technical details of the work with members of the Research and Licensing branches. It was agreed that, in general, the present scope of the project meets NRC's needs for a research study on restoration, however, the schedule and scope do not allow for a timely and detailed study of excursions, which are of immediate concern to the Licensing staff. As a consequence, the possibility of expanding this project or developing a new project is being investigated.

In order to obtain additional information on the various state regulations affecting in-situ uranium mining, visits were made to the Wyoming Department of Environmental Quality (Cheyenne), the New Mexico Environmental

Improvement Division (Sante Fe) and the Texas Departments of Water Resources and Health (Austin). From these meetings, details of specific regulations were obtained as well as an appreciation of their historical development. Publicly available environmental data on selected sites were acquired.

Task 2: Site Selection and Baseline Characterization

One of the prime requirements that must be met to achieve the goals of this project is to obtain unoxidized sediments from an ore-zone aquifer. This requires immediate preservation of the core material after it is recovered so that reactions with atmospheric oxygen are minimized. Typically, mining companies do not take adequate precautions to inhibit oxidation of their cores and, as a consequence, the material is not suitable for experimental use in this study. This necessitated finding a company that is actively drilling and was willing to allow us to take core samples from their site. Uranium Resources Inc. (URI) is currently operating the Benavides and Longorio commercial-scale in-situ leach mines in south Texas. Additionally, they have recently begun operating a pilot-scale facility at their North Platte site near Douglas, Wyoming. The company management was very willing to aid us in obtaining ore, sediment and water samples from the Benavides mine. This mine has the best set of environmental data of the three mines URI is operating. A site visit in February led to the scheduling of a sampling trip in March.

Prior to the sampling trip, detailed sampling plans were developed. These were designed as guidelines to aid in the acquisition of samples and site-specific information necessary for the laboratory experiments. Based on a typical uranium roll-front leach operation (Figure 1), it was decided that ground-water samples should be collected from each discrete water-bearing zone around the ore. Lixiviant samples would have to come from an active leach field and sediments should be cored from a leached and unleached ore zone plus the unmineralized portions of the aquifer.

Sampling at the Benavides site was carried out during the week of March 22-26. Ground-water samples were taken from the ore zone, the reduced and

FIGURE 1. In-Situ Uranium Mine Showing Injection, Recovery and Monitoring Well Pattern

oxidized portions of the aquifer immediately adjacent to the ore zone, the ore-zone aquifer a few hundred meters on each side of the ore zone, and the oxidized aquifer above the ore-zone aquifer. Lixiviant samples with high (66 ppm) and low (3 ppm) uranium content were obtained from one of the active well fields. All water samples were taken using submersible stainless steel pumps installed in fiberglass-cased wells. Galvanized piping connected the pump to the surface sampling equipment. Prior to collecting samples for analysis, the ground-water sampling wells were pumped until temperature, pH and Eh stabilized. The lixiviant samples were taken from producing extraction wells and these wells did not required purging. In addition to temperature, pH and Eh, the specific conductivity, alkalinity, dissolved oxygen content, and sulfide and ammonium concentrations were measured in the field. Samples were preserved according to accepted procedures and shipped to PNL for additional chemical analysis. Large volume samples (25L or more) were collected from selected wells for use in the laboratory experiments. Four coreholes were drilled, from which a total of approximately 59 feet of core were recovered. The material represents samples of leached and unleached ore, reduced sediments downgradient of a roll front, and the aquifer above the ore-zone aquifer. The core material was described, photographed and packaged in air-tight containers in the field.

The products of this sampling trip will provide sufficient material to perform the study on natural restoration (Task 3) and to begin the evaluation of induced restoration techniques (Task 4).

Task 3: Mobility of Potential Contaminants

The primary objective of this experimental task is the performance of laboratory studies on natural restoration using the samples collected from the aquifer environment. A laboratory test plan was written this quarter to guide the work, and laboratory space and equipment are being acquired. The experiments will begin early in the third quarter of this fiscal year. This task is also developing input to a report assessing the existing literature on restoration of uranium in-situ mine sites. This report is discussed

further in the following section.

Task 4: Restoration Techniques

Experimental work in this task is not scheduled to begin until the beginning of next fiscal year, however, during this quarter a draft experimental plan was written as a companion document to the natural restoration test plan. These experimental test plans have been used to determine sample requirements and will enable an efficient startup of laboratory work.

Progress was made on several sections of the internal report that will summarize restoration requirements, types of induced restoration, theoretical effectiveness of natural restoration and the results of restoration efforts at selected in-situ mines. The majority of the information will come from published sources and the document is intended to be used as a working tool to guide the research in this project. Because of this, the document will not be published independently, but sections of it will be used in the forthcoming topical reports and the final technical report.

Highlights

A cooperative agreement was established with Uranium Resources Incorporated (URI) to exchange data and acquire samples from their mine in south Texas. Sufficient ore, sediment, ground-water and lixiviant were collected to supply the experimental tasks. The opportunity exists for additional sampling at URI's Wyoming operation, if the need arises.

Projected Work for Next Quarter

The solutions and sediments collected at URI's Benavides mine will be analyzed and the data used in a baseline modeling study and in the experimental work. Laboratory experiments on natural restoration will commence and the literature review on restoration procedures will be completed.

Reports

No reports were completed this quarter.

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